

# COATED PARTICLE FUEL FOR HIGH TEMPERATURE GAS COOLED REACTORS

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Roy Huddle, having invented the coated particle in Harwell 1957, stated in the early 1970s that we know now everything about particles and coatings and should be going over to deal with other problems. This was on the occasion of the Dragon fuel performance information meeting London 1973: How wrong a genius be! It took until 1978 that really good particles were made in Germany, then during the Japanese HTTR production in the 1990s and finally the Chinese 2000-2001 campaign for HTR-10.

Here, we present a review of history and present status. Today, good fuel is measured by different standards from the seventies: where  $9 \cdot 10^{-4}$  initial free heavy metal fraction was typical for early AVR carbide fuel and  $3 \cdot 10^{-4}$  initial free heavy metal fraction was acceptable for oxide fuel in THTR, we insist on values more than an order of magnitude below this value today. Half a percent of particle failure at the end-of-irradiation, another ancient standard, is not even acceptable today, even for the most severe accidents. While legislation and licensing has not changed, one of the reasons we insist on these improvements is the preference for passive systems rather than active controls of earlier times.

After renewed HTGR interest, we are reporting about the start of new or reactivated coated particle work in several parts of the world, considering the aspects of designs/ traditional and new materials, manufacturing technologies/ quality control/ quality assurance, irradiation and accident performance, modeling and performance predictions, and fuel cycle aspects and spent fuel treatment.

In very general terms, the coated particle should be strong, reliable, retentive, and affordable. These properties have to be quantified and will be eventually optimized for a specific application system. Results obtained so far indicate that the same particle can be used for steam cycle applications with 700-750°C helium coolant gas exit, for gas turbine applications at 850-900°C and for process heat/hydrogen generation applications with 950°C outlet temperatures.

There is a clear set of standards for modern high quality fuel in terms of low levels of heavy metal contamination, manufacture-induced particle defects during fuel body and fuel element making, irradiation/accident induced particle failures and limits on fission product release from intact particles.

While gas-cooled reactor design is still open-ended with blocks for the prismatic and spherical fuel elements for the pebble-bed design, there is near worldwide agreement on high quality fuel: a 500  $\mu\text{m}$  diameter  $\text{UO}_2$  kernel of 10% enrichment is surrounded by a 100  $\mu\text{m}$  thick sacrificial buffer layer to be followed by a dense inner pyrocarbon layer, a high quality silicon carbide layer of 35  $\mu\text{m}$  thickness and theoretical density and another outer pyrocarbon layer. Good performance has been demonstrated both under operational and under accident conditions, i.e. to 10% FIMA and maximum 1600°C afterwards. And it is the wide-ranging demonstration experience that makes this particle superior.

Recommendations are made for further work:

1. Generation of data for presently manufactured materials, e.g. SiC strength and strength distribution, PyC creep and shrinkage and many more material data sets.
2. Renewed start of irradiation and accident testing of modern coated particle fuel.
3. Analysis of existing and newly created data with a view to demonstrate satisfactory performance at burnups beyond 10% FIMA and complete fission product retention even in accidents that go beyond 1600°C for a short period of time.

This work should proceed at both national and international level.

**KEYWORDS :** High Temperature Reactor, TRISO Coated Particle Fuel, Zirconium Carbide

## 1. INTRODUCTION

High temperature gas-cooled reactors are characterized

by an all-ceramic core, a core structure made of graphite as moderator and reflector, helium gas as a single phase inert coolant, coated particle fuel and a low power density

core. The use of refractory core materials combined with helium coolant allows high coolant temperatures up to 950°C and a high thermal efficiency results in a number of significant advantages. The low power density and large heat capacity of the graphitic core, the absence of coolant phase changes, and the prompt negative temperature coefficient represent inherent safety advantages.

One of the attractive features of the HTGR is its flexibility in the use of fuel cycles. All early HTGRs were based on the uranium-thorium cycle because of its excellent economics and resource utilization. It used high-enriched uranium (HEU) as the initial fuel and thorium as the fertile material, while the U-233 fissile material bred from the thorium could be utilized simultaneously during reactor operation or recycled for use in new fuel element production.

For the low-enriched uranium (LEU) cycle designed to minimize the chance of diverting fissile material at any point in the cycle, it was not necessary to change the design of the coated particle nor that of the reactor core. HTGR operation can be switched from one fuel cycle to another by means of the “normal” refueling schedule with only marginal adjustments. This was verified in the AVR reactor where insertion of low enriched fuel elements began in 1982. Variations of the LEU cycle include once-through or recycling of discharged fissile material.

The basic fuel containing unit is given in form of a tiny coated particle as was originally suggested and patented

by R. Huddle in 1957 and 1959, respectively. Since that time countries like Germany, the United Kingdom, France, Belgium, the United States, Russia, India, Japan, China, South Africa, and the Republic of Korea have been conducting research on coated particle fuel or producing at a large scale for the operation of HTGRs. A total of approximately 52 t of heavy metal has been used worldwide for coated particle fabrication.

The particle of ~1 mm diameter has a kernel to contain the fissile material protected by a sequence of coating layers. A reactor core for 400-600 MW(th) will contain between  $10^9$  and  $10^{10}$  individual fuel particles. The particles are embedded in a graphite matrix to form the fuel elements. Various designs have been proposed according to different purposes and extensively tested over the years. At present, the SiC TRISO coated fuel particle has been accepted by most countries as the reference concept, but R&D is continuing for further improvement and extension of the fuel envelope.

The SiC TRISO fuel particle consists of a dense heavy metal oxide or carbide spherical kernel enclosed in four successive layers:

- (1) low density pyrocarbon buffer to provide void volume for gaseous fission products, to accommodate kernel swelling, and to represent a sacrificial layer for fission fragments;
- (2) inner high density pyrocarbon, a gas-tight coating and diffusion barrier for metallic fission products to

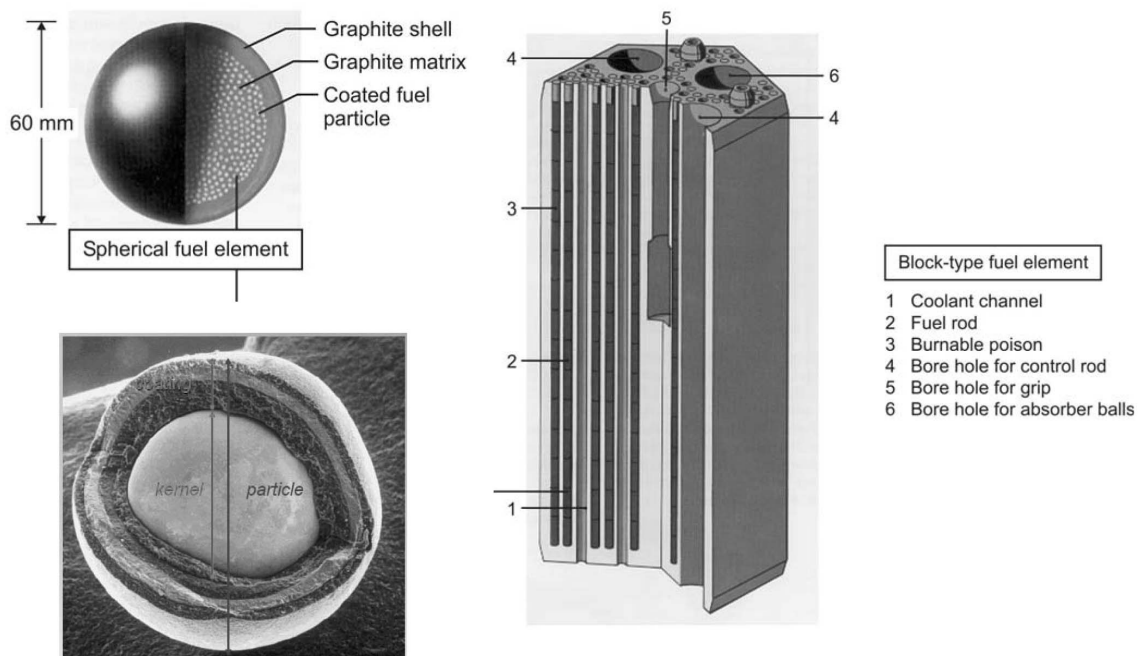


Fig. 1. Schematic of Coated Fuel Particle and Fuel Element for HTGRs

protect the kernel from  $\text{Cl}_2$  and to reduce tensile stresses on the SiC;

- (3) high density silicon carbide acting as pressure retaining layer and primary metallic fission product diffusion barrier;
- (4) outer high density pyrocarbon, a further diffusion barrier for gaseous and metallic fission products, which reduces tensile stress on the SiC, protects the SiC during particle handling and sphere/compact formation, and provides bonding surface for the overcoating.

The type of fuel element has developed in different directions (see Fig. 1):

- the spherical fuel element, composed of a 50 mm diameter fuel zone with around  $10^4$  coated particles uniformly dispersed in a graphitic matrix, surrounded by a 5 mm fuel-free carbon outer zone; this pebble bed concept was and is pursued in Germany, Russia, China, South Africa;
- cylindrical or annular fuel compact inserted in a hexagonal graphite block. The fuel block contains bore holes that are, in the US design, either coolant channels or filled with coated particles containing fuel compacts. In the Japanese design (not shown in the figure), the bore holes of a fuel assembly are filled with fuel rods ("pin-in-block") to contain the compacts, while the coolant is flowing through the annular gap between rod and inner surface of the bore hole. Somewhat different from the above was the fuel element design in the Dragon Reactor Experiment (DRE) in the United Kingdom which consisted of a cluster of six driver fuel rods with each made up from fuel compacts within graphite sleeves, and a central rod that contained an experimental section. Helium coolant was flowing through the annular gap between graphite tube and fuel body. A total of 25 fuel element varieties was developed during the course of Dragon reactor operation.

## 2. HISTORY OF COATED PARTICLE FUEL DEVELOPMENT

### 2.1 Coated Particle Development within the Dragon Project

DRAGON at Winfrith in the UK was the first demonstration high temperature gas-cooled reactor reaching initial criticality in 1964. Thirteen OECD countries supported this project to serve the growing needs of the HTGR community for irradiation testing of fuels and fuel elements, and technological tests of components and materials.

A wide variety of types of fuel particles has been developed, fabricated, tested, and inserted into the Dragon core [1]. They included kernels made of uranium monocarbide, uranium dicarbide, uranium dicarbide/thorium dicarbide, uranium monocarbide/zirconium monocarbide, or uranium

dioxide. The fuel kernels were uncoated or coated with pyrocarbon only ("emitting" type fuel), or – later – with a sequence of layers pyrocarbon/silicon carbide/ pyrocarbon ("retaining" type fuel). Furthermore process parameters during the fuel manufacture were changed to develop optimal designs.

The initial Dragon fuel was designed for the thorium cycle. For the purpose of reducing fuel power density, uranium carbide was mixed with zirconium carbide as alloying diluent to form (U,Zr)C driver fuel. For experimentation, the center rods of three fuel element assemblies were used each containing a wide variety of fuel and graphite specimens during the course of DRE operation.

In later charges, the  $\text{UO}_2\cdot 10\text{C}$  kernel with a TRISO coating was applied in the driver zone. Furthermore the particles received a thick overcoating. A significant portion of fuel compositions and coatings, and also for different irradiation times with the goal of extending fuel lifetime. They contained oxide or carbide mixed compounds, involving low-enriched uranium, thorium and plutonium, and with BISO or TRISO coatings. At a later stage, spherical fuel elements from the German programs could be irradiated in Dragon as well as pin-in-block design fuel elements or integral multi-hole graphite block fuel element segments to investigate the prismatic type fuel as was used for the US Fort St. Vrain HTGR. The Dragon project was terminated in 1975.

### 2.2 Coated Particle Design Development in Germany

The AVR in Jülich, Germany, was operated between 1966-1988, and was primarily used to test the pebble-bed concept, the spherical fuel element, and reactor internal components [2]. Fueling of the first core loading started with about 30,000 fuel elements, 70,000 moderator (graphite) balls, and 3000 absorber balls. Eventually, there were around 110,000 spheres in the system to produce a thermal power of 46 MW.

In sum, more than 290,000 spherical fuel elements of 5 different types and 15 variants (carbide/oxide, BISO/TRISO, HEU/LEU) with more than 6 billion coated fuel particles plus about 80,000 graphite (moderator) balls were inserted into the core. Most of the fuel elements were recycled several times before being discharged. The fuel element design was also changed early; starting with reload charge 3, the change was from machined graphite shells to pressed matrix materials.

Using high enriched mixed carbide/oxide fuel at the beginning, the reactor core was, since mid 1982, gradually converted to low enriched fuel. The detailed fuel composition of the reactor core over time is indicated in Fig. 2. The composition of the total reactor inventory was about 50% of HEU and 50% LEU fuel at the end of operation.

In Germany, early fuel development and testing was oriented towards the requirements of the 300 MW(e)

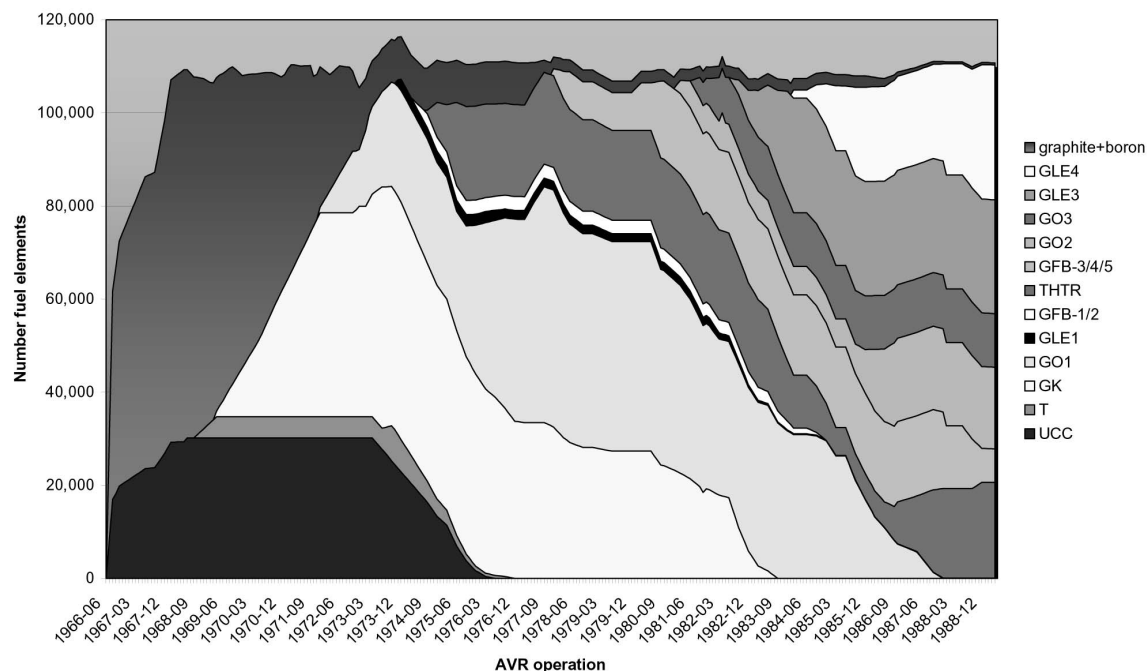


Fig. 2. Fuel Composition of AVR Core During Operation

THTR prototype and for successor reactor designs for gas turbine and process heat applications. These included – apart from the sphere as reference – also block-type fuel for the prismatic design.

Three reference particle types were suggested for the HEU/Th fuel cycle, (Th,U)O<sub>2</sub> HTI BISO, (Th,U)O<sub>2</sub> LTI TRISO, and the two-particle system with fissile UC<sub>2</sub> TRISO plus fertile ThO<sub>2</sub> TRISO. As a further development, high-density UCO kernels were considered as an alternative to the US low-density WAR (weak acid resin derived) kernels with partial conversion. One UCO (and ThO<sub>2</sub>) production charge was introduced as reload 13-3 into the AVR core.

Proliferation considerations in the late 1970s resulted in the decision to eliminate HEU fuel and to switch the fuel cycle to LEU. The LEU UO<sub>2</sub> particle with a 500 µm diameter kernel and a TRISO coating design verified during the HEU phase was made the new UO<sub>2</sub> LTI TRISO reference particle. Carbide fuel is no longer considered at all due to its complicated manufacturing process, fast diffusion of U into the PyC, and excessive release of strontium and rare earths. A first set of LEU specifications was fixed before the first large-scale production of 24,611 fuel elements were made for reload 19 in the AVR. With the shut-down of the THTR-300 reactor, all HTGR fuel fabrication activities in Germany were terminated in 1988.

## 2.3 Coated Particle Design Development in the USA

The fuel concept pursued in the USA since the beginning was based on the two-particle concept. It consisted of a

(HEU/Th)C<sub>2</sub> TRISO fissile particle and a ThC<sub>2</sub> or ThO<sub>2</sub> fertile particle. The US experimental reactor Peach Bottom I used a dense pyrocarbon coating on the kernel in the first core loading and low density/high density pyrocarbon coating combination (buffered isotropic or BISO) in the second core loading. The follow-on Fort St. Vrain plant used a TRISO coating on both fissile and fertile particles. Carbide fuel was chosen for the fissile particle, because it shows the weaker reaction with the pyrocarbon (amoeba) compared to UO<sub>2</sub> at the high temperatures and temperature gradients as given in the graphite block. The fertile particle had a larger kernel, with the resulting difference in particle size enabling mechanical separation during reprocessing.

At the end of the 1970s, the nuclear non-proliferation policies of the US government resulted in a change to LEU fissile fuel (maximum enrichment 20%). The reference fissile material since 1981 is UCO. The initial reference fertile material, ThO<sub>2</sub>, was changed to UCO or UO<sub>2</sub> in 1989. The reference two-particle system for the MHTGR concept became the 350 µm UCO or UO<sub>2</sub> with 19.9% enrichment as the fissile, and the 500 µm UCO with natural uranium as the fertile particle, both surrounded by a TRISO coating. The irradiation performance of the US fuel as was observed in the test HRB-21, however, was not good due to high failures. A series of differences were identified in a comparison between US and German fabrication methods [3]. These resulted in revised coating specifications.

Coated particle fuel production has been restarted in the USA in support of the NGNP (Next Generation Nuclear

Reactor) project and UCO fuel with a TRISO coating is currently under irradiation in the Advanced Test Reactor (ATR) in Idaho.

## 2.4 New HTGR Fuel Production Activities around the World

In recent years, various countries have started or resumed fuel manufacture activities launching extensive R&D programs with the goal of developing coated particle fuel for future HTGRs. It includes in a first step the reproduction of good-quality German fuel from the past and in a later step to expand the limits of that fuel towards higher temperatures and higher burnups.

In the beginning of the 1990s, China started a fuel R&D program focusing on the manufacture of the first core loading for the HTR-10 experimental reactor. Based on the German reference design of a spherical fuel element, China has developed fabrication technologies with which 20,000 fuel spheres have been manufactured [4]. With regard to the 500 MW(th) follow-on plant HTR-PM, design work has started to upgrade the existing fuel facility to be followed by the construction of a prototype fuel plant with a capacity of 280,000 spheres per year.

New activities in France on HTGR fuel manufacture are concentrating on the Very High Temperature Reactor (VHTR) system. A review of the experience from the past was followed by the construction of an experimental manufacturing line, CAPRI, dedicated to the production of  $\text{UO}_2$  TRISO coated particles and subsequent compacting, but flexible enough to also examine other fuel configurations [5].

Japan has long been engaged in the establishment of its own HTGR fuel production line. The fuel inserted into their 30 MW(th) block-type HTTR test reactor is SiC TRISO fuel designed for comparatively high temperatures (up to  $\sim 1500^\circ\text{C}$  design limit) and low burnup (max. 31.5 GWd/t) [6]. During the HTTR normal operation, fractional release of fission gas of Kr-88 was lower than  $1 \cdot 10^{-8}$  at full power confirming the high quality of the fuel fabricated by mass-production. New efforts are directed towards a modified fuel design with ZrC coating as an improved barrier against fission product release.

With the plans of South Africa to start construction of the PBMR in the near future, the necessary infrastructure for fuel fabrication on a large scale is being established. The main aim of the fuel production is to reproduce as much as possible the latest design of the German fuel sphere. Irradiation testing of the first fuel produced is planned for the near future [7].

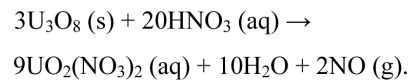
Still at an early stage, but with ambitious goals is the HTGR fuel development program in the Republic of Korea, which is part of the so-called “Nuclear Hydrogen Key Technology Development Project”, NHTD, launched in 2006. Fundamental works are currently being conducted on  $\text{UO}_2$  kernel fabrication and coating technology

accompanied by the investigation of characterization and quality control techniques [8].

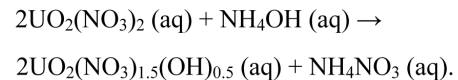
## 3. FUEL MANUFACTURE AND QUALITY CONTROL IN THE GERMAN TRADITION

### 3.1 Kernels

The manufacture of  $\text{UO}_2$  kernels is based on the external gelation process (also known as the “gel-precipitation” process). The feed material is in the form of  $\text{U}_3\text{O}_8$  powder.  $\text{U}_3\text{O}_8$  powder is dissolved in nitric acid to form a uranyl nitrate solution according to:



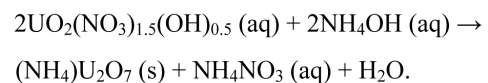
The uranyl nitrate solution is pre-neutralized with ammonium hydroxide just prior to precipitation:



A casting solution is prepared by adding small amounts of polyvinyl alcohol and tetrahydrofurfuryl alcohol to the pre-neutralized uranyl nitrate solution. This adjusts the surface tension and viscosity to ensure proper droplet formation and also assists with later uniform shrinkage and crystal growth.

Casting of microspheres is carried out in a glass column filled with the concentrated ammonium hydroxide precipitation solution. The casting solution is pneumatically fed to the nozzles at the top of the column by means of a silicone tube, where a vibrator shakes off droplets from the feed stream. 100 discrete droplets are formed per second from each nozzle. The droplets first travel through air where they attain a spherical shape as a result of surface tension. The microspheres then pass through an ammonia atmosphere, where a chemical reaction occurs with the uranyl nitrate on the surface of the microspheres causing the uranyl nitrate to precipitate as ammonium diuranate (ADU) in the outer layer of the microspheres.

The precipitated outer layer of the microspheres enables them to retain their spherical shape on impacting the precipitation solution without deforming. As the reaction continues in the casting column ammonium diuranate forms inside the microspheres, with ammonium nitrate as by product:

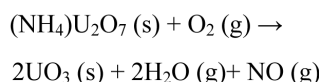


The microspheres are kept in the casting column until they are strong enough to be processed further. The diameter of a cast gel sphere is about 1.8 mm.

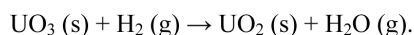
The microspheres and the accompanying precipitation solution are transferred from the casting column to a jacketed rotary flat tank. During ageing the vessel is heated with steam to 80°C. The ageing process converts fully the gel spheres to solid ammonium diuranate kernels, and ensures complete crystal growth.

After ageing the solution is drained from the vessel. The ammonium diuranate kernels in the vessel are washed with water to remove the ammonium nitrate as well as ammonium hydroxide and tetrahydrofurfuryl alcohol. Then the kernels are washed with IPA (isopropyl alcohol) to remove moisture, and any remaining ammonium nitrate, ammonium hydroxide and tetrahydrofurfuryl alcohol. The final step is to dry the kernels at 80°C under vacuum. The diameter of a dried ammonium diuranate kernel is about 1 mm and the bulk density 1 g/cm<sup>3</sup>.

After drying the ammonium diuranate kernels are calcined in air up to 430°C. The remaining organic additives are cracked and driven off during a gradual temperature increase. Above 400°C the ammonium diuranate is converted to UO<sub>3</sub>:



The diameter of a calcined kernel is about 0.8 mm and the bulk density about 2 g/cm<sup>3</sup>. After calcining follows reduction and sintering at high temperature to remove remaining impurities and densify the kernels. The process is carried out under hydrogen atmosphere to reduce the UO<sub>3</sub> to UO<sub>2</sub>:



The temperature is taken up to 1600°C in order to form dense, stoichiometric UO<sub>2</sub> kernels that have a diameter of 500 μm and a density just below the theoretical value of 10.96 g/cm<sup>3</sup>.

The final production steps are sieving to remove any under and over sized kernels, followed by sorting to remove any odd-shapes. The latter is performed on a vibrating sorting table that is slightly inclined to allow spherical kernels to roll down-hill while odd-shaped particles are vibration transported along a perpendicular direction and collected for recycling.

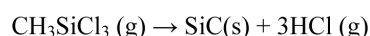
## 3.2 Manufacture of Coated Particles

The four coating layers are deposited on kernels in a heated furnace by chemical vapor deposition (CVD). Flowing gases in the furnace suspend the kernels so that

they form a fluidized bed. Gases are chosen that decompose and deposit, at temperatures up to 1600°C, certain of their constituents on the surface of the floating particles.

The process for depositing the four coating layers is as follows:

- Deposit a porous buffer layer on the kernels by the decomposition of acetylene (C<sub>2</sub>H<sub>2</sub>).
- Deposit an inner, dense layer of isotropic carbon on the porous buffer layer by the decomposition of a mixture of acetylene and propylene (C<sub>3</sub>H<sub>6</sub>).
- Deposit a dense, isotropic layer of SiC on the inner isotropic carbon layer by the decomposition of methyltrichlorosilane (CH<sub>3</sub>SiCl<sub>3</sub>), also known as MTS, according to the following reaction:



- Deposit an outer, dense layer of isotropic carbon on the SiC layer by the decomposition of acetylene and propylene (as with the inner pyrocarbon layer).

The following coating layer thicknesses were applied in the German HTGR fuel:

- buffer layer: 95 μm
- inner pyrolytic carbon layer: 40 μm
- silicon carbide layer: 35 μm
- outer pyrolytic carbon layer: 40 μm.

The final production steps are sieving to remove any under and over sized particles, followed by sorting to remove any odd-shaped particles on the inclined vibration table,

Overcoating of the particles is the final step whereby the matrix powder of the spherical fuel element is applied to the particles in a large rotating drum.

## 3.3 Quality Control and Quality Assurance

The apparatus for measuring particle diameters (and the associated volumes) for spherical particles in the size range applicable to kernels and coated particles is an automated optical particle size analyzer (PSA) with pneumatic particle transport. The method relies on the intensity dip observed by a detector when a particle passes through a light beam. For spherical particles, it is possible to achieve a linear response between an appropriately defined function of the intensity dip and the particle diameter. Calibration of the system is achieved by means of standard steel balls. Particles are pneumatically transported, separated and passed through the light beam where they are counted and measured at a rate of about 50 particles per second.

High density pyrolytic carbon is a polycrystalline graphitic material. Because each graphite crystallite inherently has anisotropic material properties, such as thermal expansion and fast neutron induced shrinkage, it is imperative to strive for isotropic crystallite orientation to have macroscopic properties that are homogeneous

and isotropic. For graphite samples, X-ray diffraction can be used to define and measure a so-called Bacon Anisotropy Factor (BAF) that directly relates to macroscopic material anisotropy. On the tiny layers of coated particles, normal X-ray diffraction is not possible. However, the intensity of reflected, polarized light differs depending on the orientation of the polarization direction relative to the crystallographic axes of the graphite crystal. Measurement of the ratio of the reflected intensities of a light beam, polarized first along one direction and then perpendicular to that direction, therefore yields an optical anisotropy factor (OAF). It can be shown that this OAF can be related in a consistent way to the BAF, which in turn relates to actual expected anisotropy and fuel performance. As a light beam can easily be focused onto a polished metallurgical section of a coated particle under a light microscope, an OAF profile across a pyrolytic carbon layer can conveniently be determined.

SiC and pyrolytic carbon densities are measured by means of suitable gradient density columns. This entails mixing two miscible liquids in a column, with densities that bracket the region of interest, in such a way that a uniform density gradient forms. To test for a particular layer, a representative test sample is extracted from the coater after completion of the layer and before the next layer is deposited. Pieces of the layer can then be cracked off and allowed to sink and settle in the column. The column is calibrated by means of standards of known density.

For  $\text{UO}_2$  kernels (density too high) and the porous buffer layer (liquid infiltration prevents accurate bulk density), the gradient column method fails and the particle size analyzer is used instead. The mass of a sample of (pre-sieved and sorted) kernels is determined accurately. The sample is then passed through the PSA and the sum of the volume of all the kernels in the sample is divided by the sample mass to yield the mean kernel density. The standard deviation follows dominantly from the spread in the volume of individual kernels.

The density of the buffer layer is determined in a similar way after subtracting the mean kernel volume from the mean total volume and using the appropriate mass values.

Although PSA analysis can be used to derive layer thickness, the method becomes increasingly imprecise for outer layers due to error propagation. To achieve good statistics of intrinsic layer variation over a large number of particles (100-200), X-ray microradiography is utilized. A single layer of particles is positioned directly on the emulsion of a high resolution photographic film (about 1  $\mu\text{m}$  resolution) and illuminated with an X-ray source approximately 300 mm away. With such an arrangement sharp projected images of layers can be achieved even with an X-ray tube that does not behave like a true point source, so that there is no need for a fine focus source. To distinguish between the buffer and the adjacent pyrolytic carbon layer, low energy X-rays are needed and the exposure must be in vacuum. Tube voltage and current are selected to give

the required contrast needed for the intended layers. The developed and mounted film is analyzed under a transmission light microscope equipped with a CCD camera. Layer thickness analysis is achieved by means of standard image processing software.

A very important test for SiC layer integrity is the burn-leach test. During the test, the graphite of the sample to be measured (loose coated particles, fuel sphere, fuel compact) is burnt in a combustion chamber (800°C in air) down to the SiC layer. The residual is treated with a nitric acid solution and the amount of dissolved uranium is analyzed. Since the SiC layer is corrosion resistant, the uranium found in the solution includes the U-content of the matrix material and the outer pyrocarbon and the U-content of particles with a defective SiC layer or with an incomplete coating. Test results are presented as the ratio of measured free uranium over the inserted uranium,  $U_{\text{free}}/U_{\text{tot}}$ , showing that these ratios are integer multiples of a coated particle inventory, meaning that the  $U_{\text{free}}$  is mainly from particles with a defective coating. The detection limit is typically at a level of  $1\text{-}3 \cdot 10^{-6}$  depending on the U-content of the sample, much lower than the U-content of a single defective coated particle.

Table 1 summarizes the achievements in HTGR fuel quality in terms of fractions of defective particles for the large-scale fuel production in Germany, Japan, and China [9]. German fuel manufacture was continuously improving in quality due to perfected tabling of kernels, particles and overcoated particles and the introduction of automated overcoating. Chinese HTGR first load fuel manufacture around 2000 for the HTR-10 was observed to have improved after the first few production runs. Japan's first fuel manufacture for the HTTR in the late 1990s showed very good results in terms of low defect fractions despite the high particle volume density of 30% (compared to below 10% for the fuel spheres).

**Table 1.** Results from German, Chinese and Japanese Burn-Leach Tests

| Manufacturer | Defect fraction     |                     |
|--------------|---------------------|---------------------|
|              | Average             | Upper 95% limit     |
| Germany      | $3.9 \cdot 10^{-5}$ | $4.6 \cdot 10^{-5}$ |
| China        | $5.1 \cdot 10^{-5}$ | $6.7 \cdot 10^{-5}$ |
| Japan        | $7.6 \cdot 10^{-5}$ | $8.1 \cdot 10^{-5}$ |

## 4. OPERATIONAL EXPERIENCE

### 4.1 Irradiation Testing

HTGR fuel has to undergo extensive irradiation testing

to demonstrate appropriate fuel performance. Also the operation of HTGR reactors is a source of valuable information. Most of these results are described in [10]. More recent irradiation testing was conducted within the frame of the Chinese fuel qualification program and of the HTR-F/F1 and RAPHAEL fuel research programs of the European Commission. As part of the fuel program in Europe, the irradiation experiments HFR-EU1 with fuel spheres from German and Chinese production, and HFR-EU1bis with German spheres have been initiated. The former is a presently on-going test, while the latter was completed in 2006. Also in the United States, the irradiation testing of newly fabricated UCO fuel has started (AGR-1) which contains fuel variants with different types of PyC and/or SiC layers. Others countries are planning the irradiation of their fuel to start in the near future.

The HFR-EU1 irradiation experiment [11] has the main objective of exploring the limits of present high-quality  $\text{UO}_2$  TRISO fuel. The three German and two Chinese fuel spheres are planned to be irradiated to a burnup of up to 20% FIMA. The irradiation temperature varies between 950°C at the surface and 1100°C in the center of the spheres; the irradiation time is 600 days. While this test is focusing on extreme burnup, the EU1bis test, simpler in its design, was to check the performance limits in terms of temperature, reaching a maximum of 1250°C in the sphere center to approach operating conditions of a VHTR. The R/B measurements of various short-lived fission gases are

given in Fig. 3. There are indications that there may have existed some manufacture-induced defective particles in the fuel balls [12]. The increasing trend later in the irradiation, in conjunction with predictions of expected release from a failed particle, indicate additional particles may have failed during irradiation. Post-irradiation examination is currently going on at the NRG in Petten, and the European Joint Research Center ITU in Karlsruhe.

## 4.2 Coated Particle Failure Mechanisms

Fuel performance is dependent upon temperature, fast fluence and burnup trajectory experienced in service, which varies spatially within the core. The modular HTGR concept, with its emphasis on passive safety features, relies upon the capability of the fuel to retain radionuclides to a very high degree during both normal operation and accident conditions. In the following sections, the major failure mechanisms for coated particles will be described [13].

### 4.2.1 Impact of Irradiation on Pyrocarbon layers

The irradiation causes the kernel to swell and the buffer layer to shrink, thereby modifying the voidage available to the gases. The shrinkage, swelling and creep behavior of the pyrocarbons is complex. Under irradiation, PyC shrinks in both the radial and tangential direction. At modest fast neutron fluences of  $2 \cdot 10^{25} \text{ m}^{-2}$  depending on the density, temperature and anisotropy of the material, it begins to

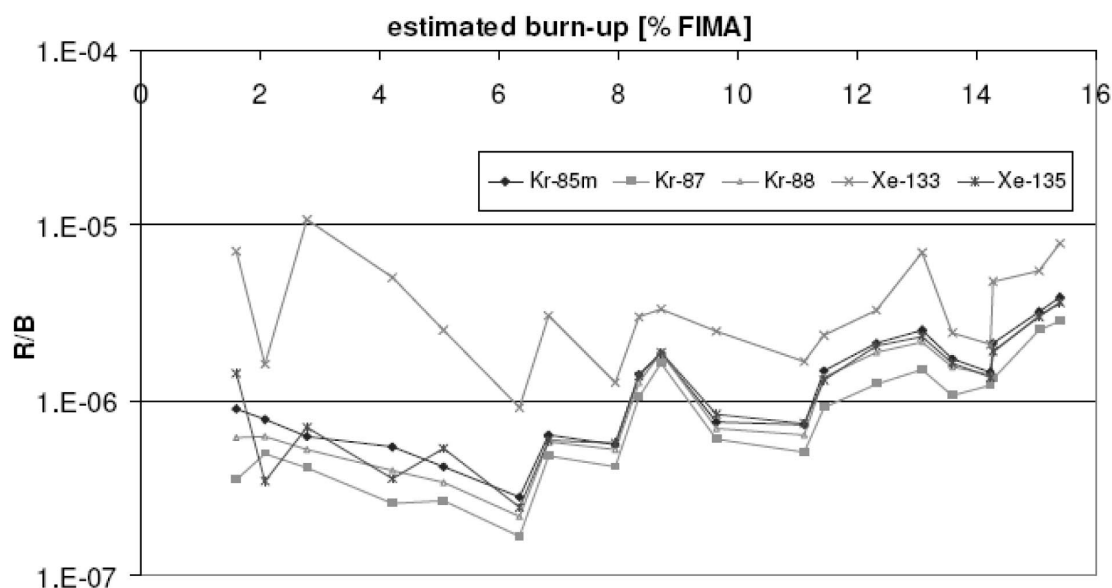


Fig. 3. HFR-EU1bis Preliminary R/B of Short-Lived Fission Gases



swell in the radial 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. This leads to a near constant PyC stress level during irradiation that relieves the tangential (or hoop) stress in the SiC layer.

In addition to irradiation-induced shrinkage, debonding at the IPyC/SiC interface has been observed in irradiation tests in the past. It refers to a detachment of weakly bonded coating layers due to tensile stresses which are generated by the PyC shrinkage under irradiation. The shrinkage observed in US irradiation testing was much larger than anticipated and led to tangential stresses in the PyC which cracked this layer and sometimes the adjacent SiC layer also. This failure mechanism has been attributed to high anisotropy in the PyC layer observed in US particles but rarely in German particles.

In AVR, one low-enriched  $\text{UO}_2$  LTI BISO reload charge of 2446 spheres (AVR 6-2) showed densification of the buffer layer that – combined with overheating from a design error (1.4 instead of 1.0 g U-235 per fuel element) – had been leading to coating cracks and subsequent massive particle failure up to 50% [14] that was observed as steep increase in AVR primary circuit R/B measurements. Therefore, this fuel variant was subsequently removed from the reactor.

#### 4.2.2 Kernel Migration

Kernel migration, i.e., the movement of the particle kernel toward the TRISO coating, also known as the amoeba effect, may lead to failure of the particle if the kernel reaches the inner PyC layer. The phenomenon is associated with carbon transport in the particle in the presence of a temperature gradient. The movement of carbon mass down the temperature gradient appears in photomicrographs of fuel as a movement of the kernel up the temperature gradient. This phenomenon is, apart from the temperature gradient in the fuel, also dependent on temperature and burnup. Kernel migration has a stronger influence in prismatic cores with  $\text{UO}_2$  fuel and usually higher particle loadings and power densities, whereas for spherical fuel elements, the effect is considered negligible (and was never observed). In oxide fuel, the amoeba effect is definitely due to the availability of free oxygen (CO). The suppression of oxygen release by means of gettering additives to the  $\text{UO}_2$  kernel was successfully demonstrated in the Dragon reactor and by others.

The spearhead attack in early Dragon fuel [15] and also observed during postirradiation examinations of AVR UCC and T type fuel elements is pyrocarbon cracking from fission fragment recoil, when there is no well-defined buffer layer and can be completely eliminated in modern fuel.

#### 4.2.3 Fission Product Attack

Past irradiation experiments indicate that fission

products can be transported from the kernel to the inner surface of the SiC where they interact and may damage or even fail the SiC layer. In older uranium carbide kernels, rare earth fission product migration was of concern, while in  $\text{UO}_2$  kernels, palladium and some of the rare earths may be an issue. In UCO kernels, the mobility of fission products is usually limited due to the generation of either carbide or oxide compounds. However, Pd transport has still been observed in UCO coated particle fuel. Also silver has been observed to migrate through apparently intact particles and be released into the reactor coolant system. The migration of the fission products is thought to be functions of time and temperature and burnup as well as temperature gradient, which means that it may play a more important role in prismatic reactors. In LEU fuels, the concentration of Ag and Pd is much greater than in HEU fuel of similar burnups due to higher yields from plutonium fissions.

CO generated during irradiation of  $\text{UO}_2$  kernels can attack the SiC layer if the inner pyrocarbon layer is either permeable or cracked. At low partial pressures of CO, SiC may be converted to  $\text{SiO}$ , thereby making the coating retention ineffective.

#### 4.2.4 Pressure Vessel Failure

During irradiation, fission gases are released from the kernel into the porous buffer layer. The inner pressure build-up from both fission gases and the CO is increasing with burnup and results in tensile stresses on the dense coating layers of the particle. Assuming the buffer to be sufficiently large and the tensile strength of the coating to be sufficiently large, particle failure may occur only, if during the coating process fabrication specifications were not reached, e.g., particles with insufficient or missing buffer layers. Irradiation experiments should ensure that this potential failure mechanism is accounted for.

## 5. MODELING OF COATED PARTICLE FUEL PERFORMANCE

### 5.1 Calculation Modeling Approaches of Coated Particle Failure

The fabrication and operation of HTGR fuel has always been accompanied by intensive efforts of mathematical modeling of the performance of the fuel taking into consideration as far as possible the physical phenomena that may occur. Modeling can assist both at the fabrication stage in identifying optimal fuel designs, and at the irradiation stage in finding appropriate irradiation conditions or postcalculating the performance of irradiated fuel, and finally at the accident simulation stage in assessing the fuel and fission product release behavior under elevated temperature conditions.

Numerous computer models have been developed varying from simple approaches such as the simplified

Booth formula to predict the release of fission gases from the particle kernel, to sophisticated numerical modeling of fission product behavior in the fuel. Most models have been described in [10] as the result of the IAEA Coordinated Research Project CRP-2, on HTGR Fuel Performance and Fission Product Behavior in GCRs.

In a follow-up IAEA project, CRP-6, which is currently on-going, the enormous progress in terms of modeling fuel performance under normal operating and accident conditions will be reported [16]. Many countries are in the stage of developing, verifying, and validating respective computer codes which model the mechanical performance of coated particles by calculating the tangential stresses within each of the three load bearing layers (IPyC, SiC, OPyC) over the course of the irradiation. It requires knowledge of various properties of the particle's constituent materials and also the way these values vary over the course of the irradiation. More experimental work on material properties is necessary to reduce uncertainties in the input data. There needs to be a continual interaction between modelers and experimenters to ensure that what is observed is being modeled.

One major task within CRP-6 is benchmarking as an important step for the validation and verification of computer models against experimental data and also against each other, thus being an ideal support for further development and/or refinement. For the exercises agreed upon in the benchmark for normal operating conditions, a total of 11 models were applied: the French ATLAS, the German codes CONVOL and PANAMA, a Japanese coated particle failure code, the Korean COPA, the Russian GOLT-V1, the South African FSAP, a Turkish Finite Element code, the UK code system STAPLE/STRESS-3, and from the US, PARFUME and a GA code. Fig. 4 gives a summary of the results obtained from these codes [17], a prediction for the irradiation experiment HFR-EU1 mentioned above.

The results exhibit an excessively broad range: PANAMA predicts the first particle to fail (which is about equivalent to reaching a failure fraction level of  $10^{-4}$ ) at a burnup between 14% FIMA ( $T_{irr} = 1100^{\circ}\text{C}$ ) and 20% FIMA ( $T_{irr} = 950^{\circ}\text{C}$ ). The GOLT-V1 results revealing a strong dependence on material properties show the first particle failure between 14 and 16% FIMA. Calculations with STRESS3 in connection with the statistical code STAPLE results in a failure fraction exceeding the level of  $10^{-4}$  (or 1 failed particle) near 14% FIMA. With PARFUME applying two options of calculating CO pressure, two irradiation temperatures and two sets of SiC strength data, the totally eight predictive calculations for the predicted failure fractions are ranging between  $4 \cdot 10^{-8}$  and 0.43. The ATLAS code using a finite element method to calculate the thermal and mechanical performance in connection with a Monte-Carlo method, the calculation of some  $10^7$  random particles resulted in the assessment of a first particle to fail, when the burnup has reached approximately 18% FIMA. Further

work will be necessary (and is planned within the national and IAEA programs) to resolve these differences. In practical terms, however, this is of no concern where fuels are irradiated to 10% FIMA and no irradiation-induced failures were predicted nor observed.

## 5.2 Open Questions

It is accepted knowledge that modeling can only be useful when the pertinent material data are well known and their applicability is guaranteed. It is therefore necessary to have coated particle material examined both prior and after irradiation. Plans to investigate material properties are being addressed by US, South African, South Korean, Chinese, and European Union fuel development programs.

The determination of the property "SiC strength" is central to all fuel particle mechanical modeling: the primary failure criterion is given by the condition when

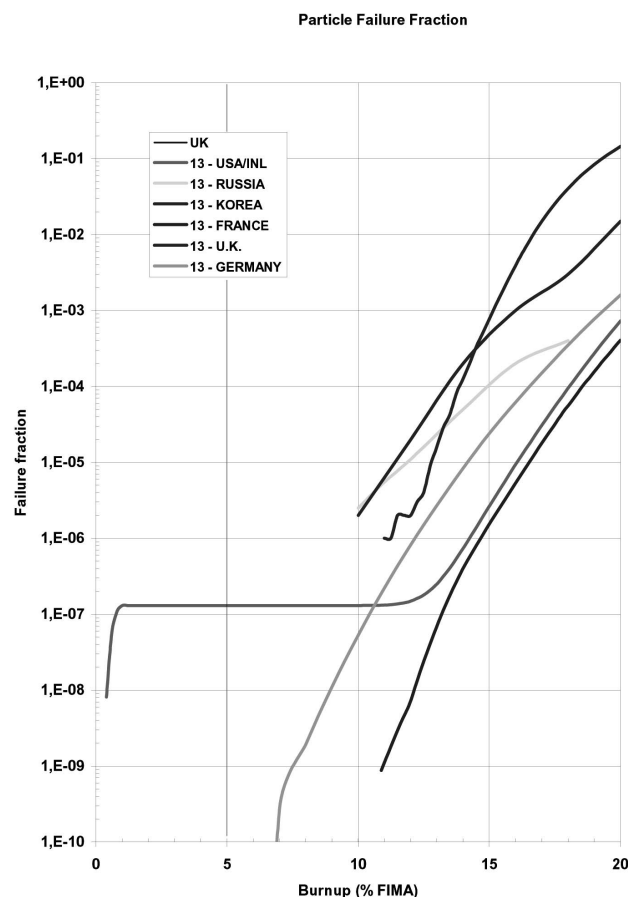


Fig. 4. Prediction of HFR-EU1 Fuel Performance with Computer Models from Different Countries

the induced stress exceeds the individual particle's SiC ultimate tensile strength. As is typical for ceramic materials, strengths are statistically distributed and it has been shown that at least 30 ring samples have to be measured [18]. In addition, this distribution is known to be influenced by irradiation under HTGR conditions. The specific problem is that SiC strength has not been measured on any of the high performance  $\text{UO}_2$  TRISO fuels. The values taken for "reference" calculations have been measured before and after (only modest) irradiation on particle batch EO 1607 from high-enriched (Th,U) $\text{O}_2$  TRISO particles manufactured in 1978.

## 6. SPENT FUEL TREATMENT

The association with a uranium/thorium fuel cycle for an HTGR was the reprocessing of the bred fissile material (U-233). In Germany, the development of economic reprocessing methods was initiated in 1966 leading to the so-called THOREX process scheme. THOREX was based on a liquid-liquid extraction method using the two immiscible liquids nitric acid as aqueous phase and a mixture of tributylphosphate and kerosene as organic phase. The process was verified at the Research Center Jülich in a semi-technical facility called JUPITER. Work in Germany on reprocessing of HTGR fuel, however, was abandoned in 1985 and rather the option of interim storage over several decades with future final disposal favored [19].

With the discussion about the VHTR as the next generation, new concepts of HTGR waste treatment strategies other than direct disposal are being developed towards a further closure of the fuel cycle, a requirement for environmentally benign nuclear energy. Recycling of spent fuel, and partitioning and transmutation of actinides and long-lived fission product species, plus the immobilization of the remainder are the steps which eventually result in a minimization of radioactive waste in connection with much shorter periods of time in the order of hundreds rather than millions of years, during which the waste represents a jeopardy to public and environment. Very high temperature reactors may play an important role in reducing the toxicity of the waste through "deep burning" of plutonium and the long-lived actinides.

The aim of transmutation is to transform long-lived, highly radiotoxic actinides by reaction with neutrons into mostly short-lived and less toxic species. This process, however, does not change heat production of the waste nor does it reduce the waste. Current (and future) R&D is concentrating on efficient methods for the necessary isotope separation and the subsequent preparation of the "new" fuel with a wide variety of potential kernel compositions, as well as the feasibility of transmutation either in specially designed nuclear reactors or in accelerator driven systems (ADS). The utilization of fuel with plutonium and minor actinides for future HTGRs is also under consideration.

## 7. COATED PARTICLE CONCEPTS FOR FUTURE HTGRS

### 7.1 Fuel Requirements in Modular Concepts

Process heat applications with HTGRs, particularly hydrogen production, typically require higher coolant and fuel temperatures. In the 1990's modular HTGR concepts began to incorporate gas turbine power conversion systems to improve plant efficiency and economics, requiring reactor outlet coolant temperatures in the range of 850 to 900°C. Much of the future interest is in application of the modular HTGR for hydrogen production using thermochemical or electrolytical water splitting processes, resulting in concepts with reactor outlet coolant temperatures of 1000°C or higher.

Direct cycle gas turbine core design analysis has been performed for the German HHT project with pebble-bed reactors and – at the conceptual design level – for two prismatic core designs, the GT-MHR in the US and the GTHTR300 in Japan. The relationship between maximum fuel temperature and coolant temperatures is generally consistent with the reactor operating experience for the fixed core reactors (Dragon and Fort. St. Vrain), with assumed reductions in core bypass flow, power peaking and flow redistribution factors by improvements in reactor design.

Conceptual design for a prismatic core thermochemical water splitting reactor is at a very early stage. The Next Generation Nuclear Plant (NGNP) project in the US has set as a goal the conditions of a 490°C inlet and a 1000°C outlet temperature of the coolant and a maximum average fuel temperature of 1250°C (SiC fuel) and 1400°C (ZrC fuel), respectively. While the former has been successfully demonstrated (with the possible exception of a potential Ag-110m problem), the latter option needs an additional massive demonstration effort. The maximum fuel temperature projections assume that further advancements in key areas (reducing core bypass flow, improved optimization of core power distribution and power/flow balancing across the core) can be made to hold the fuel temperatures to values comparable to the GT-MHR with a 150°C increase in both coolant outlet temperature and average coolant temperature rise across the core.

### 7.2 Advanced Designs with Zirconium Carbide

Advanced fuel particle concepts using ZrC as a substitute for, or in addition to SiC have been studied to a considerable degree since the 1970s. The major advantage of a ZrC coating compared to SiC is seen in its superior temperature capability in the 2000-2400°C range. Also ZrC may be more resistant to chemical corrosion by fission products, particularly by Pd that corrodes SiC. Some coated particles with ZrC layers have been shown to survive irradiation at elevated temperatures better than those with SiC [20].

Two coating designs using ZrC will be discussed:

- a particle for use at very high temperatures well above those to which SiC can be subjected, in which ZrC is used

to replace SiC in an otherwise standard TRISO particle (ZrC TRISO); and

- a more-retentive particle for use at intermediate temperatures, in which a layer of ZrC is applied over a carbon seal coat that has been applied directly to an oxide fuel kernel, followed by the standard coatings in a regular SiC-based TRISO particle. This particle with a thin ZrC coating was initially used on  $\text{UO}_2$  kernels to serve as an oxygen getter to prevent kernel migration through coatings, and it was referred to as a  $\text{UO}_2^*$  particle [21].

### 7.2.1 LEU ZrC TRISO Coated Fuel

In the course of developing a ZrC based coated particle, irradiation and post-irradiation heating tests have shown capability to retain key radionuclides (e.g., cesium) to temperatures substantially beyond the capabilities of SiC TRISO fuel. Also, considerable advances have been made in the development of reliable and practical ZrC coating processes.

In one Japanese side-by-side test of TRISO particles of both SiC and ZrC, irradiated to a burn up of 4.5% FIMA at temperatures ranging from 1400 to 1650°C, there was complete coating failure of only one of 2400 particles with ZrC, as opposed to 20 failures in the same number of particles with SiC [22]. Irradiated particles with ZrC also survive at much higher rates when subsequently exposed to high-temperature heating tests. When heated to 2400°C at 1°C per second and held for more than 1.5 hours, there was only a 1% failure of particles with ZrC, compared to 90% failure in particles with SiC by the time the temperature had reached 2400°C. ZrC retains cesium and possibly silver better than SiC.

While the higher temperature capability of ZrC TRISO fuel is well established, there are two areas of concern:

- A stable protective oxide layer is not formed to the same degree as for SiC in an oxidizing environment of air or water.
- Retention of certain fission products by SiC seems to be better than by ZrC. In particular, Ru-106 has been found to have higher diffusion rates in ZrC than in SiC, and because Pd has not been found concentrated at the inner surface of ZrC layers there is concern that this could indicate migration through the coating.

### 7.2.2 LEU SiC TRISO Coated $\text{UO}_2^*$ Fuel

A particle design with a dense pyrocarbon seal coat and thin ZrC coating applied directly to a  $\text{UO}_2$  kernel, followed by the typical SiC TRISO coatings, has been designated  $\text{UO}_2^*$ . Some such particles with a 9-14  $\mu\text{m}$  ZrC layer, along with standard SiC TRISO particles, were tested at 900°C and 1200°C (Fig. 5).

The objective of the thin ZrC layer, which was not expected to survive the irradiation, was to serve as a getter for oxygen and to retard kernel migration. Particles of each fuel type were subsequently heated for 10,000 hours at temperatures of 1200, 1350, and 1500°C. The  $\text{UO}_2^*$  fuel type was the only one that did not release a detectable amount (less than 0.01%) of any fission product in any of these post-irradiation heating tests. In addition to Cs and Ce, this included the more diffusive Ag-110m and Eu-155 isotopes for which release from individual particles for all other fuel types at 1500°C ranged as high as 15 to 100%. All  $\text{UO}_2$  TRISO particles without a ZrC layer released 100% of the silver at 1500°C, so this thin ZrC layer applied

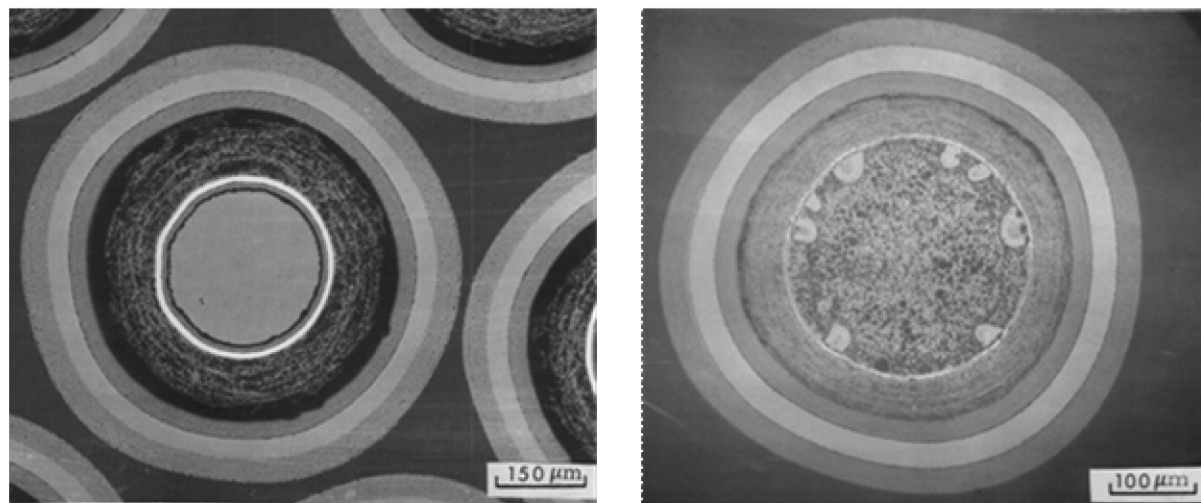


Fig. 5.  $\text{UO}_2^*$  Particles Before and After Irradiation in HRB-15B

over the  $\text{UO}_2$  kernel in  $\text{UO}_2^*$  fuel led to an improvement in retention of this and other important fission products.

Surprisingly, this thin layer of ZrC remained intact during the irradiation and X-ray examination of  $\text{UO}_2^*$  particles revealed that the ZrC layer served to restrict the swelling of the  $\text{UO}_2$  kernel to about 1%, whereas the same kernels in regular TRISO particles expanded by 10.6%. Also the effect of considerable improvement of fission product retention capability suggests the possibility of other benefits in which the presence of the ZrC layer either led to improvements in SiC layers by protecting them during the coating process, or by protecting them until eventual ZrC failure during irradiation reduced releases, or both.

The observation that none of the regular  $\text{UO}_2$  particles that released silver at 100% (and europium at 16%) gave any evidence of Ru-106 release at 1500°C, whereas ZrC TRISO particles at 1600°C for 4500 hours indicated release rates of about 10% for Ru-106, brings again into question the ability of ZrC TRISO particles to be used at high temperatures. More testing of  $\text{UO}_2^*$  fuel would be needed to evaluate its full potential.

## 8. CONCLUSIONS

A large body of international experience with coated particle fuels for HTGRs, beginning in the 1950s, has led to a convergence on a LEU oxide SiC LTI TRISO particle with some variation in kernel composition, diameter and coating thicknesses depending on the performance objectives. As a result, the failure mechanisms and performance limits as a function of particle design parameters are relatively well established. Continuing improvements in fuel performance and fission product transport models are further increasing the understanding of the behavior and limitations of this particle design. HTGR gas turbine and process heat applications under development require a significant increase in fuel temperatures relative to the steam cycle applications developed earlier. While the LEU SiC TRISO fuel system performance capability is sufficient to support initial deployment of these applications, achievement of their full economic potential may be enhanced by the development of advanced fuels with higher temperature capability.

Failure probabilities are functions of temperature, burnup, fluence and temperature gradient in the particle and details of the particle design. Based on the previous German experience, TRISO coated fuel is usually designed, manufactured, and operating conditions are constrained such that none of the fuel failure mechanisms are expected to be significant. Fission product releases during irradiation and heatup testing will be dominated by pre-existing as-manufactured defects in the production fuel and heavy metal contamination outside of the SiC layer and initially defective particles. Strict process control and proper statistical quality control is used to limit as-manufactured defects in coated particle fuel.

Because of the potentially improved fission product retention of  $\text{UO}_2^*$  TRISO particles with thin ZrC layers applied directly over  $\text{UO}_2$  kernels, it seems highly desirable to conduct irradiation tests with a more significant number of particles and at higher temperatures, followed by post-irradiation heating tests to demonstrate accident performance.

National engagement as well as bilateral, multi-national, European-wide and world-wide (IAEA) cooperation in HTR fuel development is ongoing and is expected to further improve fuel performance and the ability to make reliable predictions.

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