NUCLEAR FUELS STATUS

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This morning I am going to talk to you about coated particle fuel performance from a modular High Temperature Gas Reactor (HTGR). The experimental results I am going to talk about came from the Oak Ridge National Laboratory, in cooperation with some of our foreign partners. The talk is directed to the results from the HTGR program, the commercial program and the HGR programs, so the temperature range is much lower.

First of all, (see Figure 1), I would like to speak about the fuel particle concept: The functional requirements, the performance limiting mechanisms and the temperature range we are looking at. All of these give you an up-to-date view of what our fuel performance is at normal operating conditions (when temperatures are less than 1250 C), the results we can expect at the accident conditions (testing temperatures greater than 1250 C, up to 2500 C), and techniques for performance characterization.

The HTGR, or the gas cooled program, fuel particle provides two specific functions (see Figure 2). One is a source of fissile material. The other is the primary containment system for fission products. The fuel source is either a dense oxide or carbide, an oxicarbide, uranium, thorium, plutonium, or a mix of two. We looked at the actual oxicarbon compound, as well as a solid state mixture.

The containment is a dense ceramic coating formed by Chemical Vapor Deposition (CVD) and deposition in fluidized bed coatings (see Figure 3). It has two primary coatings of BISO (see Figure 2). In our program, we looked at two pyrocarbon coatings. We also looked at some silicon pyrocarbon mixtures with the outer layer. The reference design is a TRISO coating (Figure 2), four layer design: two pyrocarbons followed by silicon carbide, in some cases, and an outer carbon-carbon layer.

Figure 3 gives a quick view of what I am talking about here. It is a Scanning Electron Microscope (SEM) photo of a particle we purposely broke. The fuel kernel is surrounded by four layers; the buffer layer, the inner PyC silicon barrier and an outer PyC.

In order to crack the particle, we used a small micrometer device. It was an intentional break of an underradiated fuel. We wanted to look at the coatings.

For the modular HTGR, heavy reliance is placed on the coated particle as a containment concept to prevent fission product release (see Figure 4). Fission products are kept at the site of their origin. Particle/containment performance can be continually monitored in the reactor. The high quality fuel is a requirement. On the average we require a quality level

of 6 equivalent failed particle, per 100,000. We distribute the containment system over the entire population of the fuel, rather than a few barriers.

I indicated earlier that the UCO kernel is our reference: a solid gel mixture derived of UO_2 (see Figure 5). It has an enrichment of 20 percent. Normal operating conditions are a temperature range from 750 to 1250 degrees C. I think anything beyond that is considered accident temperatures. Burnup is 26 percent FIMA. Fast fluence is less than 5, and we are talking about fairly low power levels. We are talking about 150 milliwatts for these designs. We have testing at higher power levels, and higher orders of magnitude show that UCO fuel is superior to UO_2 and UC_2 under similar operating conditions. That's why it was selected as a reference.

After a number of years of in-reactor testing, we have identified the basic or dominant fuel performance limiting mechanisms for our fuel (see Figure 6). They are pressure vessel failures; meaning the internal pressures exceed the strength. The silicon carbide layer had massive failures, and we had a lot of fission product release. We also had silicon carbide coating failure. This did not necessarily cause massive releases but it did contribute to synergistic effects.

The dominant mechanics of kernel migration and carbon transport, in the presence of a thermal gradient, results ultimately in kernel/silicon carbide contact and layer degradation. In this mechanism, fission products migrate through the silicon carbide layer and interact. Again, this results in layer degradation. Consequently, pressure vessel is not of standard requirement.

Thermal dissociation is the decomposition of the silicon-carbide layer resulting in loss of coating integrity. It is active above 1600 degrees C for various periods of time. Finally, we have fast neutron damage, causing differential expansion/contraction of the pyrocarbon layer, cracks in the layer and a complete loss of coating integrity.

The thermal migration data has been around for a number of years. We have looked at different fuel kernel designs with respect to the envelope that we could allow. We found with HTGR that if the UO_2 fuels fall within the envelope at temperatures 1200-1300 degrees C, we can not use that design for HTGR applications (see Figure 6). UC_2 falls somewhat below but fairly close to oxide. Basically, what we ultimately came up with in a kernel design was the UC-O concept. This concept eliminates the thermal migration problem as well as the fission product release problem from the UC_2 fuels.

Data about thermal decomposition was attained in the accident testing program. At 2500 degrees C, the silicon carbide coating disappears. It's primarily a carbon, grain coating. Interestingly enough, this particle has not failed catastrophically. Instead it has expanded. The coating is still visually intact; fission products have been lost to a fairly large degree. There was a burn up of about 3 percent and loss of about 25 percent of the cesium. Fission gasses will be lost at somewhat less than the 25 percent level.

We demonstrated in the German program that we can fabricate fuel with very high levels of quality. They produced hundreds of kilograms of material using production scale facilities. The U.S. did it with prototypic modular scale facilities.

In-reactor fuel failure levels have been demonstrated less than 10^{-4}. In fact, the level is about 3×10^{-5} with very high confidence, and that is based on fission gas release data. Temperatures go up to 1200 degrees C, 12 percent FIMA and relatively high fast fluence.

Accident conditions for the German program are temperatures that temperatures range between 1250 and 2500 degrees C. The U.S. program uses any condition that causes the fuel to be less than 1600 degrees C (see Figure 8).

In accident simulation test, at 1600 degrees C for periods up to 500 hours, no significant fission product release was observed. This is based primarily upon the German data. From that we can show, with very high confidence, the induced failure levels of 10_{-5} range. At 1800 degrees C and above, we do find some significant amounts of metallic fission products being released after short periods (Short period are in hours not minutes). In a ramp test where we took fuel to 2500 degree C in 50 hours, no detectable fission products were released.

Figure 9 shows ramp heating data. You can see a plot of the fraction release krypton 58 as a function of heating time. At 1600 degrees C we see that these levels are 10^{-6} level compared to what near 10_{-4} would be for a single particle failure. 1800 degrees C we see that after periods of some 50 hours or so we start seeing degradation of the fuel. Basically this is a diffusion of fission products through the silicon carbide. At 2100 degrees C you can see failure rapidly occurs.

The ramp test in Figure 10 with German data shows that you are not seeing serious silicon carbide degradation until you get to 2100 degrees C. Then you get a fairly rapid rise. This data has been used quite a few times. I think it is wrong to conclude that you can run in at 1900 degrees C or 2000 degrees for a long time. That is not true. These are only ramp test data. They need to be compared to the isothermal data, at least for our concept.

I am trying to put together a comparison of the performance attributes from the HTGR to NTP for what I knew prior to coming to this meeting (see Figure 11). I could change that quite a bit after yesterday, so it's good education for me. We have an UCO in the U.S. and UO_2 in Germany. Our coating is basically a silicon carbide TRISO design. NTP looks at zirconium carbide fuel form. The HTGR concepts is that of a machine graphite prismatic block or a sphere.

Enrichments may be the same. For the civilian program, we have about 20 percent. For the NPR program, we have fully enriched material. The Germans have 8 to 10 percent, and they have a large database on fully enriched material. This appears appropriate for NTP.

The big difference is the power produced per particle. The HTGR is a low power per particle. The power density in the core is very low. We are looking at 100 to 150 milliwatts in NTP, a watt or two per particle, maybe even beyond burnup. NTP may be, with respect to an open cycle, a tenth of a percent as indicated.

With respect to fuel quality, we demand a very high quality. I think that someone is going to say the same thing with respect to this application here. I heard things about dumping fission products out the back but that's not something to be decided by me.

Let me finalize this with what is available to us for characterization of fuel performance and fission products (see Figure 12). We have a full range of testing irradiation available to us. We also have hyper or thermal spectrum. We can achieve up to 5 watts of power per particle while maintaining in-reactor surveillance. With a full range of Post Irradiation Evaluation (PIE) capabilities, we can look at the physical metal and fission gas retention on a particle basis. We have high temperature PIE and furnaces that will go up to the HTGR program's 2000 degrees C limit. The furnaces are probably about 2800 degrees C. And we have capabilities for modeling fuel particle behavior and fission product transfer.

Outline

* *Particle Fuel Concept*
* *Functional Requirements*
* *Performance Limiting Mechanisms*
* *Fuel Performance*
    - *Normal Operation [<1250°C]*
    - *Accident Conditions [>1250°C]*
* *Methods/Techniques for*
    Characterizing Performance

Figure 1

THE COATED PARTICLE FUEL CONCEPT PROVIDES TWO BASIC
FUNCTIONS: (1) SOURCE OF FISSILE MATERIAL; AND
(2) PRIMARY CONTAINMENT FOR FISSION PRODUCTS

Fuel Source: A dense oxide, carbide, or oxi-carbide spherical
                 kernel of uranium, thorium, plutonium, or a mixture

Examples - ThO_2, UO_2, $(Th,U)O_2$, $(U, Pu)O_2$
             ThC_2, UC, UC_2, $(Th,U)C_2$
             UC_xO_y, $UO_2 + UC_2$ mixture

Containment: Dense ceramic coatings surrounding spherical fuel
                 kernel formed in succession by chemical vapor
                 deposition (CVD)

Examples -   BISO Coating, two layer design [PyC/PyC]
             TRISO COating, four layer design [PyC/PyC/SiC/PyC]

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Figure 2

BASIC FUEL UNIT OF THE MHTGR IS
THE COATED PARTICLE

SOURCE OF FISSILE FUEL MATERIAL
PRIMARY FISSION PRODUCT CONTAINMENT
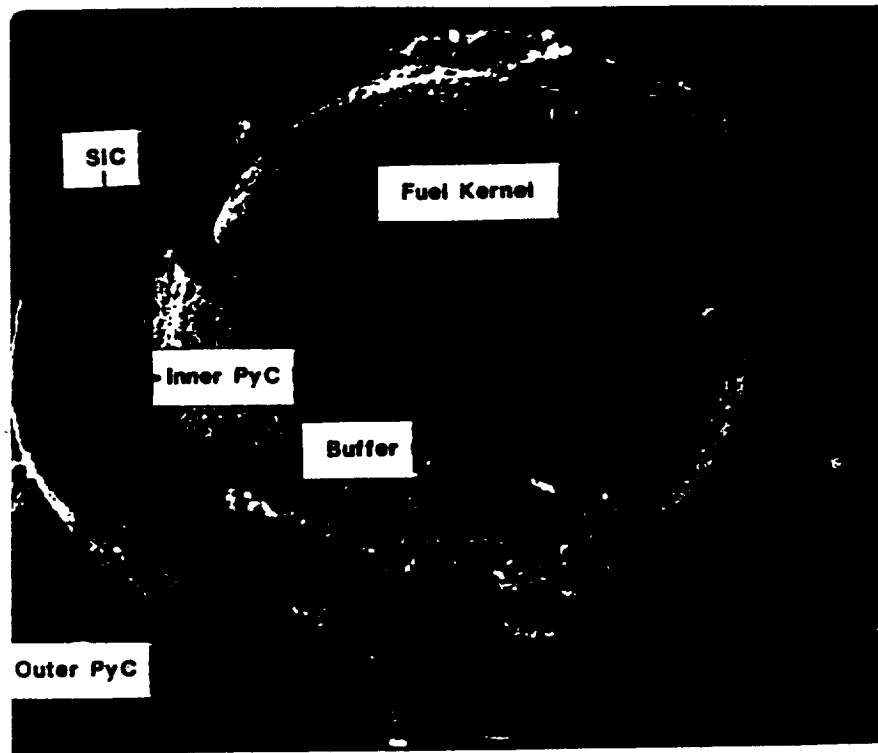

SIC

Fuel Kernel

Inner PyC

Buffer

Outer PyC

Figure 3

FOR THE MODULAR HTGR HEAVY RELIANCE IS PLACED
ON THE COATED PARTICLE CONTAINMENT
CONCEPT TO PREVENT FISSION
PRODUCT RELEASE

o   Fission Products are kept at the site of their origin
    under normal and off-normal events.

o   Particle/containment performance can be continually
    monitored in-reactor by measuring primary circuit
    activity.

o   High Quality fuel fabrication by requirement - on
    average, quality level of < 6 equivalent failed
    particle per 100,000.

o   Containment system distributed over 10^{10} microspheres,
    rather than depending upon only a few barriers.

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Figure 4

TRISO-COATED UCO SELECTED AS REFERENCE FISSILE PARTICLE FOR MODULAR HTGR BASED ON ITS FABRICABILITY AND FISSION PRODUCT RETENTION CAPABILITIES

o UCO Kernel is a Sol-Gel mixture of UO_2 (80%) and UC_2 (20%) with a 20% enrichment.

o Normal Operating Conditions:

Temperature (°C)	750 - 1250
Burnup (% FIMA)	≤ 26
Fast Fluence ($10^{25} n/m^2$)	≤ 5
Power/particle (mW)	~ 150

o Performance superior to UO_2 and UC_2 under similar operating conditions.

Figure 5

EXTENSIVE IN-REACTOR TESTING AND POSTIRRADIATION EXAMINATION TESTS HAVE IDENTIFIED DOMINATE FUEL PARTICLE PERFORMANCE LIMITING MECHANISMS

1. Pressure Vessel Failure - SiC tensile stress induced by internal gas pressure exceeds layer strength resulting in total coating failure and massive FP release

2. SiC Coating Failure [Contributes to synergistic effects]
   o Kernel Migration - carbon transport in presence of thermal gradient results ultimately in kernel/SiC contact and layer degradation,
   o FP Interaction - FPs released from kernel diffuse to SiC, chemically interact resulting in layer degradation,
   o Thermal Disassociation - decomposition of SiC layer resulting in loss of coating integrity, active above 1600·C.

3. Fast Neutron Damage - differential expansion/contraction of pyrocarbon layer resulting in loss of coating integrity.
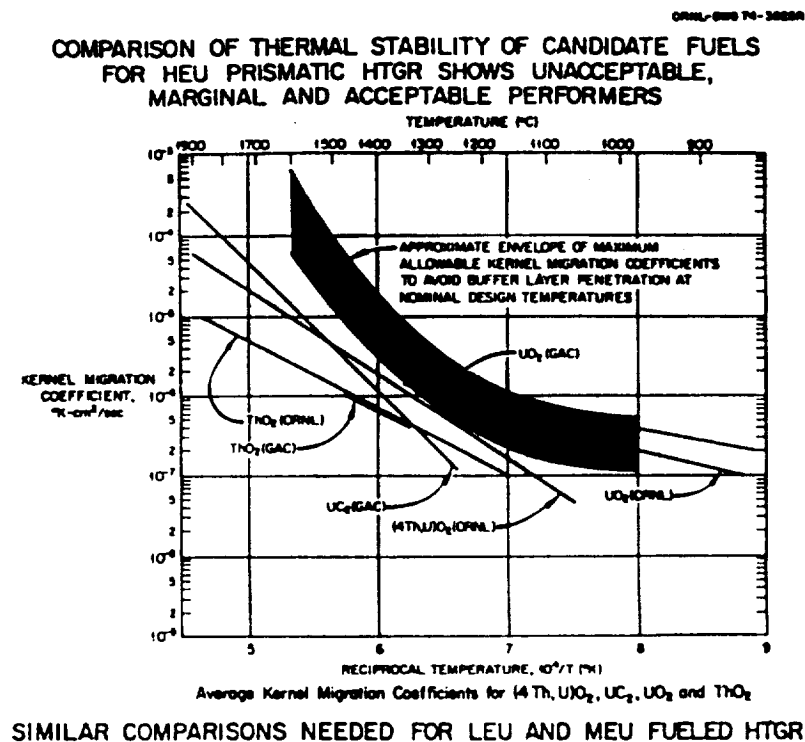
Figure 6

COMPARISON OF THERMAL STABILITY OF CANDIDATE FUELS
FOR HEU PRISMATIC HTGR SHOWS UNACCEPTABLE,
MARGINAL AND ACCEPTABLE PERFORMERS

Average Kernel Migration Coefficients for (4 Th, U)O₂, UC₂, UO₂ and ThO₂

SIMILAR COMPARISONS NEEDED FOR LEU AND MEU FUELED HTGR

Figure 7

COATED PARTICLE FUEL PERFORMANCE
(Modular HTGR)

Accident Conditions [Temperatures > 1250·C]

o Modular HTGR design limits maximum fuel temperatures
to < 1600·C under all conditions.

o In accident simulation tests at 1600·C for periods up
to 500 h, no significant FP release was observed.

o For accident simulation tests at 1800·C and above,
significant FP release (gaseous/metallic) observed
after short periods.

o Ramp tests of 50 h duration to 2500·C, exhibited no
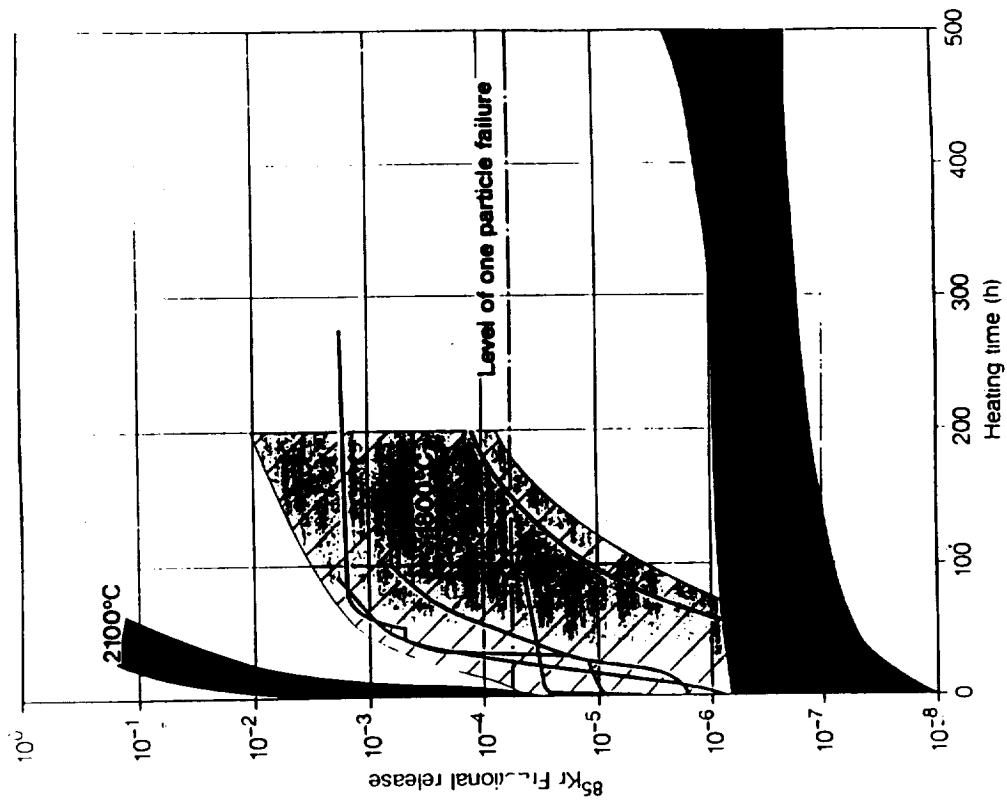detectable FP release beyond HTGR peak accident conditions.
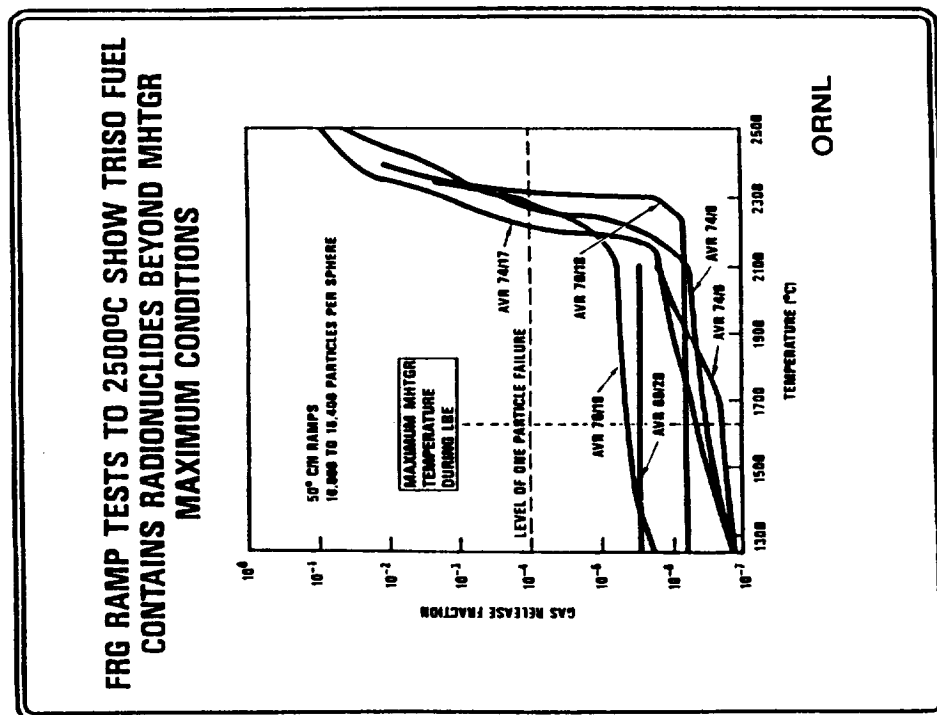
Figure 8

Figure 9

FRG RAMP TESTS TO 2500°C SHOW TRISO FUEL CONTAINS RADIONUCLIDES BEYOND MHTGR MAXIMUM CONDITIONS

ORNL

Figure 10

Comparison of Coated Particle Performance Attributes
(HTGR and NTP Concepts)

Attribute	Modular HTGR		NTP
	US	FRG	
Particle Design – Kernel – Coating	UCO SiC TRISO	UO2 SiC TRISO	[UC,UC2,?] [ZrC TRISO]
Fuel Form	Prisimatic	Sphere	[Unbonded or Compacts]
Enrichment (% U–235) *	< 20 / >90	8 to 10	[> 90]
Power/Particle (mW)	< 150	< 100	[1000 to 2000]
Burnup [% FIMA] *	< 26 / < 75	< 12	[< 25]
Operating Temperature (°C) – Normal – Accident	750 to 1250 < 1600	700 to 1200 < 1600	[> 1250] [?]
Fuel Quality	< 6 E-5	< 6 E-5	[?]

* Cuꞩ⨪ MHTGR /UPR-MHTGR

Figure 11

SPECIALIZED TECHNIQUES AND METHODS ARE AVAILABLE AT ORNL TO CHARACTERIZE COATED PARTICLE PERFORMANCE AND FISSION PRODUCT BEHAVIOR

o Irradiation Testing
 - Accelerated environment
 - Thermal Spectrum with
   Spectral Tailoring
 - High Power, up to 5W/
   particle
 - In-Reactor Surveillance

o Postirradiation Examination (PIE)
 - Metrology/Ceramography
 - Fission Metal Retention/Particle
 - Fission Gas Retention/Particle
 - Electron Microscopy/Microprobe

o High Temperature PIE
 - Remote Furnaces
   Temperatures up to 2000·C
 - Quantitative FP Release
   Determination
 - Post Test Characterization

o Modeling/Documentation
 - Fuel Particle Behavior
 - Fission Product Release/Transport
 - Statistical Analysis
 - Performance Assessments

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Figure 12