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Performance modeling of Deep Burn TRISO fuel using ZrC as a load-bearing layer and an oxygen getter

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ABSTRACT

The effects of design choices for the TRISO particle fuel were explored in order to determine their contribution to attaining high-burnup in Deep Burn modular helium reactor fuels containing transuranics from light water reactor spent fuel. The new design features were: (1) ZrC coating substituted for the SiC, allowing the fuel to survive higher accident temperatures; (2) pyrocarbon/SiC "alloy" substituted for the inner pyrocarbon coating to reduce layer failure and (3) pyrocarbon seal coat and thin ZrC oxygen getter coating on the kernel to eliminate CO. Fuel performance was evaluated using General Atomics Company's PISA code. The only acceptable design has a 200-μm kernel diameter coupled with at least 150-μm thick, 50% porosity buffer, a 15-μm ZrC getter over a 10-μm pyrocarbon seal coat on the kernel, an alloy inner pyrocarbon, and ZrC substituted for SiC. The code predicted that during a 1600 °C postulated accident at 70% FIMA, the ZrC failure probability is $<10^{-4}$.

1. Introduction

The Deep Burn reactor proposed by the General Atomics Company [1] utilizes reprocessed spent nuclear fuel from light water reactors. Recovered plutonium (mostly ^{239}Pu) and other minor transuranics are fabricated into TRISO fuel particles. ^{238}U, which typically accounts for at least 95% of the spent fuel mass, and a miniscule amount of ^{235}U and ^{236}U separated from the spent fuel are to be reused in LWRs. For a one-pass Deep Burn cycle in a 600 MW_t Gas-Turbine Modular Helium Reactor (GT-MHR) operating at 1000 °C, the transuranic materials are irradiated to a very high maximum burnup of 70% FIMA (fission per initial metal atom). The spent fuel is sent directly to the repository. This exceptionally high-burnup is achievable because of the special properties of the coated particle fuel employing ZrC.

Deep Burn meets the three objectives of the nuclear industry and the new GNEP initiative: (1) efficient utilization of energy in spent fuel; (2) reduction in waste volume and (3) proliferation resistance (essentially no fissile material in the fuel sent to the repository).

The current design of SiC-coated TRISO fuel particles used in GT-MHR has a spherical oxide kernel containing fissile materials. The kernel is fabricated by a sol–gel technique. A typical size of the fissile kernel is 200–500 μm in diameter. A CVD-deposited buffer layer (50% dense pyrolytic carbon) surrounding the kernel provides free space for fission gases and for kernel expansion due to solid fission product swelling and fission gas bubble formation. Fission recoils are stopped in the layer. The buffer condenses during the course of irradiation. A typical buffer thickness is 100 μm. An inner pyrocarbon layer (IPyC) is CVD-deposited on top of the buffer layer. Although this layer confines fission gases, most of solid fission products can easily diffuse through it. Due to the crystal structure of pyrolytic carbon, it shrinks under fast neutron irradiation. The magnitudes of the shrinkages on the tangential and radial directions depend on the layer isotropy, density, temperature, and fast neutron fluence. The PyC shrinkage in the tangential direction (which is parallel to the SiC deposition plane) places a compressive load on the SiC while the counteracting effect of fission-gas pressure adds a tensile stress to it. A typical IPyC thickness is 35 μm. An intact high density IPyC also protects the kernel from chlorine attack, as chlorine-bearing gas is used to deposit SiC.

A dense, CVD-deposited ceramic SiC layer on top of the IPyC functions as a main load-bearing layer and as a fission product containment. An intact SiC coat retains both gaseous and metallic fission products except ^{110m}Ag, which can pass through at temperatures above 1000 °C. A typical SiC thickness is 35 μm. The outermost layer is a CVD-deposited outer pyrolytic carbon (OPyC). Its main role is to isolate SiC from the surroundings, and when it shrinks, it compresses the SiC layer.

To fabricate a fuel compact, fuel particles are mixed together with a graphite binder, formed into a cylinder 12.7 mm in diameter and 50.8 mm long. Then, it is fired at high temperature to carbonize

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Table 1
Elemental composition of the Deep Burn kernel (oxides).

Element	Atom (%)
Pu	85
Np	5
Am	9
Cm	1

the binder. Failure of SiC during compact manufacturing due to particle-to-particle contact is reduced significantly by pre-applying an over-coating layer – another layer of the graphite binder – to the particles.

2. Evaluating the new design features

Each of the new design features was evaluated using a fuel performance code. Evaluations were made on:

- Conventional TRISO.
- ZrC substituted for SiC (ZrC-coated TRISO).
- ZrC getter on kernel with
  - o Conventional TRISO.
  - o ZrC-coated TRISO.
- Alloy IPyC with
  - o Normal TRISO.
  - o ZrC-coated TRISO.
  - o Conventional TRISO with ZrC getter on kernel.
  - o ZrC-coated TRISO with ZrC getter on kernel.

For all cases, a steady-state temperature of 1000 °C[1] from the BOL to the maximum EOL burnup of 70% FIMA, reaching a fast fluence of 4×10^{25} n/m^2, was assumed. A postulated accident scenario causing the fuel temperature to reach 1600 °C three days after the EOL burnup was assumed. A constant temperature gradient of 10^4 K/m across the fuel particle[2] was used during the irradiation period. Material properties of the particle fuel were assumed unchanged with fast neutron fluence, include during the accident. To allow comparison of all the cases on the same basis, a kernel diameter of 200 μm, a buffer thickness of 100 μm, an IPyC (or alloy PyC) thickness of 35 μm, an SiC (or ZrC) thickness of 35 μm, and an OPyC thickness of 40 μm were assumed. A PyC seal coat thickness of 10 μm and a ZrC getter thickness of 15 μm[3] were assumed for designs employing the ZrC scavenger. Fabrication defect of SiC (and ZrC) was taken to be 10^{-5}.

Literature search revealed the volumetric kernel swelling due to solid fission products to be as low as 0.35% [2] and as high as 1% [3] per % FIMA. Although the values were for UO_2, they were assumed to apply to the Deep Burn kernel composing mostly of plutonium oxide. For conservative estimates, the volumetric swelling of 1% per % FIMA was adopted. Kernel swelling increased fission-gas pressure because of the reduction in the in-particle free volume.

3. In-particle gas pressure

3.1. Fission-gas pressure

Fission-gas pressure as function of burnup and temperature was calculated and manually input. The total yield of Xe and Kr was 27.2%, calculated by weighting the yields from ^{239}Pu and ^{241}Am obtained from Ref. [4] by their corresponding atomic fractions in the kernel shown in Table 1.

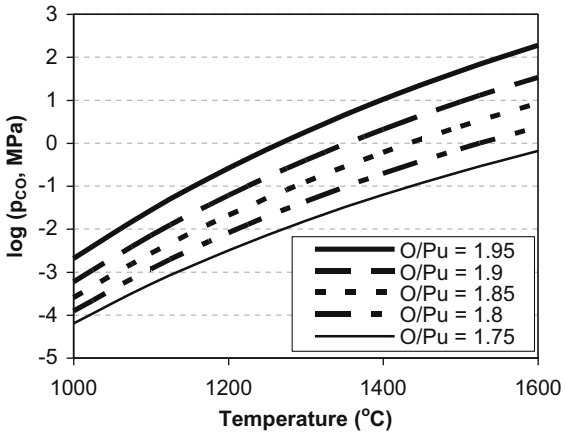

Fig. 1. Predicted CO pressure under different temperatures and O/Pu ratios.

All fission gas atoms were assumed to leave the kernel and stay in the buffer porosity, for conservative estimates. This is not the case in reality, however, as micro and macro fission-gas bubbles of various sizes form in the kernel, and as a portion of the gas atoms remains atomically dispersed in the kernel matrix. For the purpose of calculating fission-gas pressure, the buffer was taken to be 50% dense and remained 50% dense through the end of accident. Although buffer densification does occur, the total free space in the buffer layer (less that taken up by the swollen kernel) should not change substantially.

3.2. CO pressure

Following the worst-case scenario for Deep Burn, a 1600 °C accident at 70% FIMA, the carbon monoxide pressure[4] would be very large. The effect of temperature and O/Pu ratio on the CO pressure was studied and is demonstrated in Fig. 1. The required governing equation for the oxygen potential of PuO_{2-x}[5] as a function of oxygen stoichiometry and temperature was adopted from Lindemer's work [4]. The standard-state Gibb's formation energy of the $O_2 + 2C \Leftrightarrow 2CO$ reaction required to establish the CO pressure was obtained from Ref. [4]. Note that the CO pressure is thermodynamically-controlled. It does not depend on the in-particle free volume. At 70% FIMA and 1600 °C, the O/Pu ratio becomes very high, most likely reaching at least 1.92,[6] corresponding to a CO pressure in excess of 60 MPa. Fission-gas pressure calculated in the preceding section augments the CO pressure by a minimum of tens of MPa, depending on the kernel size and buffer thickness, making the overall internal gas pressure close to 100 MPa.

3.3. Americium pressure

Americium vaporization from metallic and oxide systems was investigated by Lindemer [4]. According to the Ellingham diagram of the Am-O-C system [4], at 1600 °C, Am gas contributes the largest partial pressure among the Am-containing gaseous species (Am, AmO, and AmO_2). However, at 1600 °C, the pressure is always

[1] A typical average fuel temperature. Although peak fuel temperatures for VHTRs are typically 1200-1250 °C, it will be shown later that the kernel size, the buffer thickness and the ZrC getter, not the steady-state temperature, are the main parameters in determining the fuel failure during the postulated EOL accident.

[2] A typical value based on General Atomics Company's literature.

[3] The thickness of ZrC required to consume all of the oxygen anticipated released from fission up to the EOL burnup is only a few microns. The proposed PyC seal coat and ZrC getter thicknesses accommodate fabrication, since a coating layer of only a few microns thick would not be practical to fabricate reliably.

[4] Fissioning of uranium or plutonium oxide liberates oxygen atoms. A portion of them combines with fission products to form stable fission product oxides. The remaining unbound oxygen diffuses out of the kernel and combines with carbon in the buffer layer to form the CO gas.

[5] PuO_{2-x} is a conventional short-hand notation for a mixture of PuO_2 and Pu_2O_3. For example, $PuO_{1.68}$ represents a mixture of 0.36 mol of PuO_2 and 0.32 mol of Pu_2O_3.

[6] From interpolation of entries in Table 6 of Ref. [4].