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Thermo-Mechanical Response of a TRISO Fuel Particle in a Fusion/Fission Engine for Incineration of Weapons Grade Plutonium

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Abstract - *The Laser Inertial Fusion-based (LIFE) engine is an advanced energy concept under development at Lawrence Livermore National Laboratory (LLNL). LIFE engine could be used to drive a subcritical fission blanket with fertile or fissile fuel. Current LIFE engine designs envisages fuel in pebble bed form with TRISO (tristructural isotropic) particles embedded in a graphite matrix, and pebbles flowing in molten salt Flibe ($2LiF+BeF_2$) coolant at T~700C. Weapons-grade plutonium (WGPu) fuel is an attractive option for LIFE engine involving the achievement of high fractional burnups in a short lifetime frame. However, WGPu LIFE engine operating conditions of high neutron fast fluence, high radiation damage, and high Helium and Hydrogen production pose severe challenges for typical TRISO particles. The thermo-mechanical fuel performance code HUPPCO (High burn-Up fuel Pebble Performance COde) currently under development accounts for spatial and time dependence of the material elastic properties, temperature, and irradiation swelling and creep mechanisms. In this work, some aspects of the thermo-mechanical response of TRISO particles used for incineration of weapons grade fuel in LIFE engine are analyzed. Preliminary results show the importance of developing reliable high-fidelity models of the performance of these new fuel designs and the need of new experimental data relevant to WGPu LIFE conditions.*

I. INTRODUCTION

The Laser Inertial Fusion-based (LIFE) engine is a laser-based inertial confinement fusion engine that could be used to drive a subcritical fission blanket. Various LIFE engines are been considered capable of burning fertile or fissile nuclear material, including natural or depleted uranium (DU), weapons-grade plutonium (WGPu) and spent nuclear fuel (SNF); for a detailed description see References [1-4]. In the WGPu LIFE engine, 14 MeV fusion neutrons pass the first wall and enter a Beryllium neutron multiplier region surrounded by a spherical WGPu fuel blanket cooled with Flibe molten salt.

The fission fuel blanket, in pebble bed form, contains ~ 7 metric tons of WGPu filling a region which extends ~ 76 cm in thickness. The WGPu LIFE basecase scenario assumes the fuel blanket consists of ~ 13.2 million fuel pebbles that are ~2 cm in diameter, with each fuel pebble containing ~ 2444 TRISO-coated particles embedded in a graphite matrix. TRISO particles have a Plutonium-oxycarbide based fuel kernel at their center. The basecase WGPu TRISO assumes a kernel ~300 μm in radius with ~ 13mol% PuCO and ~87 mol% ZrC. The kernel is surrounded by a ~ 102 μm thick porous (~ 56%) carbon-based buffer layer accommodates internal gas buildup. The thick buffer is followed by a three-layer coating composed of an inner pyrolytic-carbon (IPyC) layer, silicon carbide (SiC), and outer pyrolytic carbon (OPyC). In this work, WGPu TRISO PyC density is assumed to be ~ 1.90 g/cc with IPyC and OPyC ~ 30 μm and ~ 20 μm in thickness, respectively. A thick ~ 60 μm SiC layer serves as a pressure vessel for the particle. The failure of the TRISO-coated particles depends on the SiC layer remaining intact during the whole irradiation. Therefore, SiC layer structural integrity is critical to ensure fission product retention.

WGPu LIFE system neutronics and burnup analysis [4] show that WGPu LIFE fuel reaches ~99.97% fissions per initial metal atoms (FIMA) after ~9.4 years of irradiation, or end of design life (EOL), after a neutron exposure to a high fast fluence F(E > 0.1 MeV) ~3.6×10^{22} n/cm^2. After ~ 4 years of constant power (~ 3800 MWth) the ~76% FIMA burnup level has already been attained and almost all the ^{239}Pu (~ 90%) has already been destroyed. As the irradiation proceeds, gaseous fission products induce increasing tensile stresses with preassures reaching ~ 30 MPa at EOL. Note that only Xe and Kr contribute to the inner pressure, with CO and CO_2 gas production in the kernel assumed to be negligible.

At these high burnup values, stress induced in the coating layers due to fission product buildup and PyC coating dimensional changes are important. Also, WGPu LIFE engine operating conditions of high neutron fast fluence, high radiation damage, and high Helium and Hydrogen production pose severe challenges for TRISO materials. As we shall see in what follows, the feasibility of using TRISO-coated particles in LIFE WGPu engine will depend on the coating materials behavior under irradiation and their thermo-mechanical performance.

II. RADIATION EFFECTS IN TRISO COATING LAYERS

Damage and gas production in WGPu TRISO is high and is expected to affect materials behavior under irradiation (strength, swelling, and other material properties). Displacements per atom (DPA) and He and H gas production rates in C and Si were calculated [5,6] using SPECTER code [7] with its self-contained data libraries and the continuously evolving neutron spectra in the WGPu fuel blanket. The WGPu fuel blanket neutron spectrum evolution is followed over the irradiation time period in intervals of 30 days. Damage and gas production are calculated after each interval of time and added. Calculations assume that all the gas produced during a given time interval accumulates on top of the one already produced without interval relaxation. Results at EOL reported in Table 1 correspond to the total cumulative sum.

Table 1: Radiation effects in C and Si in WGPu LIFE fuel blanket after 99.97% FIMA (~9.4 years).

	C	Si
dpa	31	48
He appm	1073	351
H appm	0	624

II. MODELING THE THERMO-MECHANICAL RESPONSE OF TRISO LAYERS

To analyze the thermomechanical/chemical response of different fuel designs and help assess their performance in LIFE engine fuel blanket we are developing a new analytical stress code HUPPCO (High Burn-Up Fuel Pebble Performance Code). A preliminary version of HUPPCO is utilized in this work to perform the thermomechanical analysis of LIFE WGPu TRISO fuel. HUPPCO calculates the stress obtained from the solutions to the radial and tangential stresses, σ_r and σ_t, from the following set of constitutive equations:

$$\frac{\partial \varepsilon_r}{\partial t} = \frac{1}{E_r}\left(\frac{\partial \sigma_r}{\partial t} - 2\mu \frac{\partial \sigma_t}{\partial t}\right) + c_{irr}(\sigma_r - 2\nu\sigma_t) + c_{th}(\sigma_r - 2\nu\sigma_t)e^{-A/T_0} + \dot{S}_r(r,t) + \alpha_r \dot{T}(r,t)$$

$$\frac{\partial \varepsilon_t}{\partial t} = \frac{1}{E_t}\left((1-\mu)\frac{\partial \sigma_t}{\partial t} - \mu \frac{\partial \sigma_r}{\partial t}\right) + c_{irr}((1-\nu)\sigma_t - \nu\sigma_r) + c_{th}((1-\nu)\sigma_t - \nu\sigma_r)e^{-A/T_0} + \dot{S}_t(r,t) + \alpha_t \dot{T}(r,t)$$

$$\varepsilon_r = \frac{\partial u}{\partial r}$$

$$\varepsilon_t = \frac{u}{r} \qquad (1)$$

where ε is the strain, σ is the stress, and E, μ, and ν are, respectively, the modulus of elasticity, Poisson's ratio and Poisson's ratio in creep. Here, c_{irr} and c_{th} are the irradiation and thermal creep constants according to the formulation of the creep term adopted by Miller *et al.* [8], $\dot{S}$ is the volumetric swelling rate, t is the time, $\dot{T}$ is the temperature rate, and α is the thermal expansion coefficient. Equations (1) are formulated assuming spherical geometry and the subindices r and t refer to the radial and tangential components, respectively. Closure of the system of equations (1) is achieved by considering the elastic equilibrium equation $\nabla \cdot \sigma = 0$, which for an axisymmetric solid is given by:

$$\frac{\partial \sigma_r}{\partial r} + \frac{2}{r}\left(\sigma_r - \sigma_t\right) = 0 \qquad (2)$$

The stress-strain relations are solved analytically assuming a power series solution in time and reducing the equations to a solvable p.d.e. recursion relation for its coefficients.

III. PRELIMINARY RESULTS

In the present preliminary analysis of the stress developed in WGPu TRISO coating layers thermomechanical input parameters corresponding to specifications given in the IAEA Benchmark Case 12 [9] are adopted, i.e. PyC modulus of elasticity (E = 3.96 x 10^4 MPa), Poisson's ratio (0.33), Poisson's ratio in creep (0.5), and coefficient of thermal expansion (5.5x10^{-6} 1/K) . Input parameters for SiC are: modulus of elasticity (E = 3.70 x 10^5 MPa), Poisson's ratio (0.13), and coefficient of thermal expansion (4.9x10^{-6} 1/K). The temperature of the particle outer layer is assumed to be that of Flibe coolant, i.e. T ~ 700 C.

A polynomial dependence is used to describe the PyC radial and tangential strain-rate behavior as a function of fast fluence F given in units of F(E > 0.18 MeV) x 10^{25} n/m^2:

$\dot{S}r$ = 4.73756 x 10^{-4} F^3 - 3.80252 x 10^{-3} F^2 +1.64999 x 10^{-2} F -2.13483 x 10^{-2}

$\dot{S}t$ = -1.03249 x 10^{-3} F^3 + 5.47396 x 10^{-3} F^2 -3.29740 x 10^{-3} F -1.83549 x 10^{-2}

Irradiation creep rate for PyC is assumed to be directly proportional to both stress and fast flux. The creep constant K equal to 0.85 10^{-4} (MPa 10^{25} n/m^2)$^{-1}$ is evaluated following the expresion obtained from the analysis of CEGA database and given in [10] which assumes that K is independent of fast neutron fluence and depends on temperature and PyC density.

Our results show SiC carbide remains always under compression during the whole irradiation with large maximum negative tangential stress of ~ -460 MPa occuring at about ~ 0.6 x 10^{25} n/m^2 and tangential stress decreasing to ~ -200 MPa at 1x 10^{26} n/m^2. Tangential stresses in SiC seem to grow very slowly beyond ~ 1 x 10^{26} n/m^2; indicating a possible long survival of the particle from this perspective. However, as shown in Fig. 1, tangential stresses in PyC have attain a large maximum of ~ 600 MPa at ~ 0.6 x 10^{25} n/m^2 which might lead to PyC coating failure at early times.

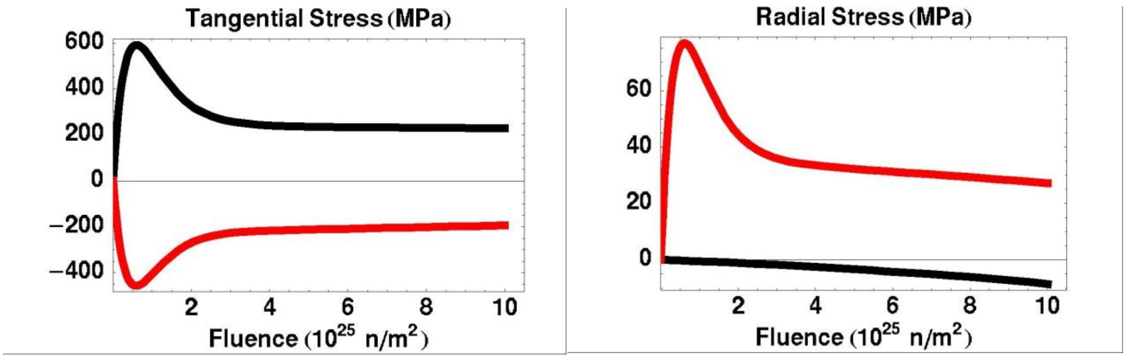

Fig. 1. Tangential and radial stress at the inner surface of PyC (black) and SiC (red) layers in WGPu TRISO. SiC layer remains under compression during the whole irradiation.

Radial stress in SiC layer never become zero or negative. The radial stress in SiC reaches ~ 75 MPa at ~ 0.6 x 10^{25} n/m^2, and then decreases to ~ 25 MPa at ~1 x 10^{26} n/m^2. Concerning PyC radial stresses Fig. 1 shows that as the pressure increases the stresses in PyC match the behavior of the inner pressure reaching ~ -7 MPa at ~ 1 x 10^{26} n/m^2.

Studies on an optimized WGPu TRISO in LIFE engine accounting for LIFE engine environment and radiation damage effects and using a HUPPCO code version that includes time and temperature dependent thermomechanical input parameters are currently underway [11].

IV. CONCLUSIONS

Our work represents a first step in the development of modeling capabilities to describe the thermo-mechanical response of WGPu TRISO under irradiation in LIFE engine. We have found that there is a wide variability for most of the important thermal - mechanical properties for pyrolytic carbon. This calls for an experimental program that will help achieve accurate modeling of TRISO particle behavior under irradiation, by accurate measurement of TRISO materials properties in representative environment. Further work is planned on systematic parameter variations and sensitivity analysis of the fuel performance to define an optimized fuel particle design and to feed new input to new LIFE engine fuel blanket concepts.

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