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Performance of Coated Particle Fuel in a Thorium Molten Salt Reactor with Solid Fuel

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Abstract – *In a completely new program, Shanghai Institute of Applied Physics, Chinese Academy of Sciences (SINAP, CAS) plans to develop a Solid Fuel Thorium Molten Salt Reactor (TMSR-SF). TMSR-SF is a 10MW, fluoride molten salt (2LiF-BeF_2) cooled experimental reactor with graphitic spherical fuel elements containing TRISO coated particles. The 60 mm diameter spherical fuel element will serve for the first core of TMSR-SF. The coated particle has a long history starting with highly releasing fuels in the early sixties where a carbonaceous coating was only added to be able to handle the early carbide fuels. The development quickly changed to oxide fuels, including both 400 µm diameter HEU (Th, U)O_2 and 500 µm diameter LEU UO_2. In this paper, we will introduce the preliminary design of TMSR-SF, and investigate the performance of spherical fuel elements under normal operating and accident conditions. Although, low failure fraction and fission products release fraction were expected, the compatibility of spherical fuel elements and molten salt is a big issue to be verified for TMSR-SF, and will be reported later.*

ACRONYMS

TRISO	Tristructural-isotropic
HEU	Highly Enriched Uranium
LEU	Low Enriched Uranium
SiC	Silicon Carbide
UO_2	Uranium Dioxide
(Th, U)O_2	Thorium-Uranium Dioxide
CO	Carbon Monoxide
PyC	Pyrolytic Carbon
IPyC	Inner Pyrolytic Carbon
OPyC	Outer Pyrolytic Carbon
EDN	Equivalent Dido Nickel
EOL	End of Life

I. INTRODUCTION

As one of the fourth generation reactors, the Molten Salt Reactor (MSR) has great prospects. The investigation for MSR can be dated back to 1940's as part of United States program to develop nuclear powered airplane. A small reactor, the Aircraft Reactor Experiment (ARE) was built at Oak Ridge to investigate the feasibility of molten fluoride fuels for aircraft propulsion reactors [1]. In 1954, the ARE was operated successfully for 9 days at a steady-state outlet temperatures ranging up to 1133 K and power up to 2.5 MWt. With the research process, the design of Molten Salt Reactor Experiment (MSRE) was begun in 1960 and the maximum power was about 8 MWt [2]. Construction of the MSRE began in 1962, and it achieved first criticality in 1965. Sustained operation at full power began in December 1966.

Since 2011, Shanghai Institute of Applied Physics, Chinese Academy of Sciences (SINAP, CAS) carried out a completely new program to develop a Solid Fuel Thorium Molten Salt Reactor (TMSR-SF) with graphitic spherical fuel elements containing TRISO coated particles. The 60 mm diameter spherical fuel element will serve for the first core of TMSR-SF, which is a 10MW, fluoride molten salt (2LiF-BeF_2) cooled experimental reactor.

In the development of High Temperature Reactors (HTRs), the coated particle has a long history starting with highly releasing fuels in the early sixties where a carbonaceous coating was only added to be able to handle the early carbide fuels. The development quickly changed to oxide fuels, both 400 µm dia. HEU (Th,U)O_2 and 500 µm dia. LEU UO_2. The dense kernels are surrounded by a ~100 µm thick buffer layer to provide free volume for the fission gases and sacrificial materials to stop fission fragments. The ultimate achievement for mechanical stability and complete fission product retention is the TRISO coating layers consisting of a 40 µm inner pyrocarbon, a 35 µm thick SiC, and a 40 µm outer pyrocarbon layer. These layers have been perfected over the last decades and provide high quality fuels, both in compacts for prismatic HTRs and in spherical fuel elements for pebble-bed HTRs.

The 60 mm diameter spherical fuel element consists of a matrix graphite that has both excellent corrosion resistance and high stability during irradiation. Its inner fuel zone contains between 10,000 and 15,000 TRISO particles, while the outermost 4-6 mm thick region are completely fuel free. The spheres are manufactured by cold isostatic pressing. This necessitates a high quality matrix graphite overcoating of the TRISO particles to avoid particle crushing during sphere pressing. TRISO particles have been qualified in a large number of irradiation tests and accident condition of heating tests.

This paper will introduce the coated particle failure mechanism within the framework of Pressure Vessel Model and the behavior of fission products release, respectively. Then, the inner pressure, stress of coating layers including both the contribution of inner pressure and the effect of irradiation shrinkage and creep of PyC, failure fraction as well as the fission products release fraction are calculated and analyzed. Based on these results, performance of TRISO fuel serviced in TMSR-SF is evaluated.

II. FUEL PERFORMANCE MODELS

II.A. Failure Mechanism

As shown in Fig. 1, the fuel kernel is surrounded by a porous pyrolytic carbon buffer, an inner dense pyrolytic carbon (IPyC) layer, a SiC layer, and an outer dense pyrolytic (OPyC) layer, sequentially. During irradiation, fission gases are released from the kernel into the porous buffer layer. Besides, the free oxygen liberated by UO_2 as uranium undergoes fission will react with carbonaceous buffer to form CO. Both fission gas and CO built up the internal gas pressure, which increases with burnup and result in tensile stresses on the dense coating layers of the particle. If these stresses exceed the tensile strength of the coating, the layer will fail.

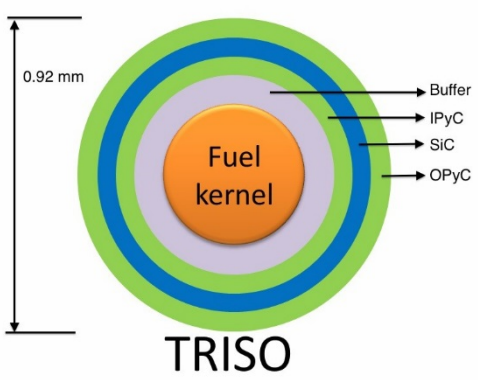

Fig. 1: Schematic diagram of TRISO coated particle

In the pressure vessel model, the SiC layer represents the wall of a simplified pressure vessel with a thin shell similar to a soap bubble. All other layers are ignored. This pressure vessel is assumed to fail immediately when the stress induced in the SiC layer exceeds the tensile strength of the SiC. In the mean time, irradiation will weaken the SiC layer, so that its strength will decrease as a function of the fast neutron fluence. The pressure inside the SiC 'vessel' is dependent on various parameters such as the yield of stable fission gases, burnup, free oxygen, and, of course, the temperature.

The probability for a pressure vessel failure can be described, in its general form, according to the following equation:

$$\Phi_{\text{total}} = 1 - \exp\left[-\ln 2 \left(\frac{\sigma_t}{\sigma_0}\right)^m\right] \qquad \text{(eq.1)}$$

where σ_0 is the tensile strength of unirradiated SiC, which is the stress corresponding to a 50% failure probability; σ_t is the stress induced in SiC layer due to the inner pressure; and m is the Weibull modulus.

In addition, since the irradiation swelling and thermal creep of PyC might influence its integrity, it is necessary to take account of the stress of PyC during the irradiation. Where, the stress due to irradiation swelling and creep of PyC can be evaluated as it is handled in Ref. [3], that is, radial stress of SiC resulted from irradiation swelling and creep of PyC can be written as:

$$\sigma_r^{SC}(r_i) = \frac{2}{c} \frac{(W-1)(\dot{S_r}+2\dot{S_t})+3X(\dot{S_r}-\dot{S_t})}{3[2W(2\nu-1)-(1+\nu)]} \qquad \text{(eq.2)}$$

for the IPyC ($r_i = r_3$), then $W = \left(\frac{r_2}{r_3}\right)^{-3}$, and

$X = \ln(\frac{r_2}{r_3})$; for the OPyC ($r_i = r_4$), then $W = \left(\frac{r_5}{r_4}\right)^{-3}$, and $X = \ln(\frac{r_5}{r_4})$. Where c is creep constant depending on temperature and fast fluence, ν is the Poisson ratio in irradiation creep, $\dot{S}_r$ and $\dot{S}_t$ are the differentials for the fast fluence induced dimensional changes in radial and tangential direction respect to fast fluence. The radial and tangential irradiation swelling rate can be written as [4]:

$$\dot{S}_r = -0.176\exp(-1.75\Gamma)$$
$$\dot{S}_t = -0.036\exp(-2.1\Gamma) - 0.01 \quad \text{(eq. 3)}$$

Where the creep constant of PyC is a function of its density and irradiation temperature, it reads:

$$C = C_0(T)[1 + 2.38(1.9 - \rho)],\ 1 < \rho < 2$$
$$C_0(T) = 1.996 \times 10^{-29} - 4.415 \times 10^{-32}T + 3.6544 \times 10^{-35}T^2 \quad \text{(eq. 4)}$$

II.B. Fission Products Release

The transport of any metallic fission or activation product could be calculated if the transport parameters of the nuclide were available. But it is impractical to evaluate the production, transport, and release of all species generated in a nuclear reactor. Actually, only few significant nuclides are selected to be analyzed depending on fission yield, transport and release properties, and radiological hazard level. Here, we focus on the long-lived nuclides strontium (^{90}Sr), silver (^{110m}Ag), cesium (^{137}Cs), and the short-lived nuclide iodine (^{131}I).

Fission products formed in the kernel have to be transported through all the coating layers before diffusing through the matrix and being released into the coolant. For fission products produced in kernel, two mechanisms dominate their releasing. One is the direct recoil, and the other is diffusion.

As for direct recoil, due to high kinetic energy, parts of the fission products formed near the kernel surface get into the buffer directly. The resulting fractional release is independent of irradiation parameters, particularly of temperature, and can be derived from purely geometrical considerations. The recoil contribution to release fraction can be calculated as [5]:

$$F_{recoil} = \frac{3}{4}\frac{d}{r_k}\left[1 - \frac{1}{12}\left(\frac{d}{r_k}\right)^2\right] \quad \text{(eq. 5)}$$

Where F_{recoil} is the recoil fraction, r_k is the radius of the kernel, and d is the mean recoil distance.

The transport of fission products through the fuel kernel is modeled as a transient diffusion process. Fick's law is employed to represent the transport process through fuel materials, hence, in spherical coordinates:

$$\frac{\partial c(r,t)}{\partial t} = D(r,t)\frac{1}{r}\frac{\partial^2(r\,c(r,t))}{\partial r^2} - \lambda c(r,t) + p(t) \quad \text{(eq. 6)}$$

where λ is the decay constant, p is the fission product production rate, c is the concentration, D is the diffusion coefficients, r is the position, and t is time. The equation is typically solved numerically with appropriate boundary and interface conditions.

During accident condition, the neutron flux is stopped and the fuel is not under irradiation anymore. So the release of long-lived or stable fission products is dominantly controlled by source-free diffusion process, which means p=0. Then, the diffusion equation can be written as:

$$\frac{\partial c}{\partial t} = D\frac{1}{r}\frac{\partial^2}{\partial r^2}(rc) \quad \text{(eq. 7)}$$

As for fission products release in coating layer, the initial condition is c=0，λ=0 for stable (or, by approximation, long-lived) fission products. Boundary condition at the outer surface $r = r_s$ is c = 0 constantly.

Ⅲ. RESULTS AND DISCUSSION

Ⅲ. A. Input Parameter

The main specification parameters of spherical fuel elements and TRISO particles for TMSR-SF are listed in Table 1, and the prospective service conditions are presented in Table 2.

Table 1: Parameters of fuel modeling of TMSR-SF TRISO fuel.

Category	Parameters	Nominal value
Fuel element characteristics	Geometry	Spherical
	Radius (cm)	3
	^{235}U enrichment (wt%)	17
	Uranium loading (g)	7
Particle geometry	Kernel diameter (μm)	500
	Buffer thickness (μm)	95
	IPyC/OPyC thickness (μm)	40
	SiC thickness (μm)	35
Material properties	Buffer density (g/cm^3)	≤1.10
	IPyC/OPyC density (g/cm^3)	1.9
	SiC density (g/cm^3)	≥3.18
	SiC Weibull modulus	8.02 [7]
	SiC strength (MPa)	834 [7]
	IPyC/OPyC Poisson ratio in irradiation creep	0.5 [4]

Table 2: Nominal and boundary irradiation conditions.

	Nominal Value	Lower bound	Upper bound

EFPD(Days)	250	-	-
Fast Fluence (10^{25}n/m^2, EDN)	0.36	-	-
Burn-up(% FIMA)	< 3	-	-
Irradiation temperature (^{o}C)	706	600	1430

///. B. Internal Gas Pressure

Both the gaseous fission products and carbon monoxide built up during irradiation will push the coating layers outward with increasing burnup. Fig.2 illustrates the inner pressure variation according to the burnup and temperature in case of TMSR-SF. The inner pressure increases with the increasing temperature and burnup. However, it is obvious that even if the temperature is up to 1430^{o}C, the inner pressure will not exceed 100 MPa.

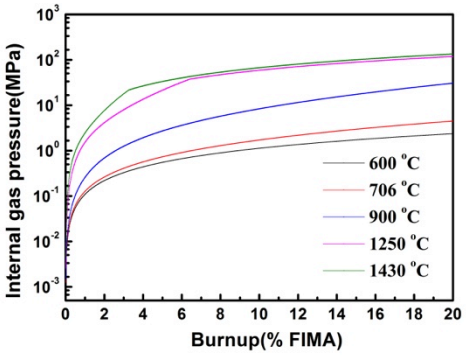

Fig. 2: Internal gas pressure of TMSR-SF particles.

///. C. Stress Level

The radial stress of SiC layer due to irradiation swelling and creep of PyC can be also estimated following the model as described in Ref. [3], the corresponding results are presented in Fig. 3. Where the creep constant was used corresponding to the irradiation temperature is 706 ^{o}C. It is indicated that at early stage of irradiation, both IPyC and OPyC push the SiC layer in compression due to their irradiation shrinkage. And, the compressive stress increases with increasing burnup. However the stress induced by OPyC starts to decrease when the burnup reaches about 5% FIMA (amount to fast neutron fluence 0.68×10^{25} n/m^2(EDN)).

Since the irradiation shrinkage and creep will generate stress in SiC and PyC layers during the irradiation. The failure distribution density of PyC abide Weibull distribution, than it reads [6]:

$$f_{PyC} = 1 - \exp\left[-\left(\frac{\sigma_{tc}}{\sigma_{med}}\right)^{m_c}\right] \qquad \text{(eq.7)}$$

Where σ_{tc} stands for the tangential stress of PyC layer, σ_{med} represents the mean strength of PyC, and m_c corresponding to the Weibull modulus. Here, we adopted the parameters as used in Ref. [7]: $\sigma_{med} = 200$ MPa, $m_c = 5$. Based on the stress calculation, the failure distribution density of IPyC is shown in Fig. 4. It is found that the failure distribution density is around 8×10^{-3} when the burnup reaches 3% FIMA. From this point, it is considered that the IPyC layer keeps intact in TMSR-SF service conditions.

In the case of TMSR-SF, the EOL burnup is less than 3% FIMA, which means both the IPyC and OPyC making compressive stress on SiC. By contrast, the internal gas pressure pulls SiC layer in tension. As presented in Fig. 5, when the burnup is less than 5% FIMA, the compressive stress in SiC due to PyC is much higher than the tensile stress generated by internal gas pressure corresponding to various irradiation temperature. It means that the stress in SiC resulted from PyC will compensate the inner pressure to a certain degree.

///. D. Failure Probability

For conservative estimation, the stress induced by PyC was neglected during the failure probability calculation. Given irradiation temperature, with the increasing of burnup, the failure probability of particles in TMSR-SF is presented in Fig. 6. It is obviously, under the circumstance of TMSR-SF, the failure probability of TRISO fuel is extremely low.

The failure fraction is also predicted for the nominal accident condition as shown in Fig. 7 (irradiation temperature 706^{o}C, 250 EFPD). Since both the operation temperature and the accident temperature are quite low comparing to the case in gas cooled reactor, the failure fraction does not increase obviously after 100 hours accident.

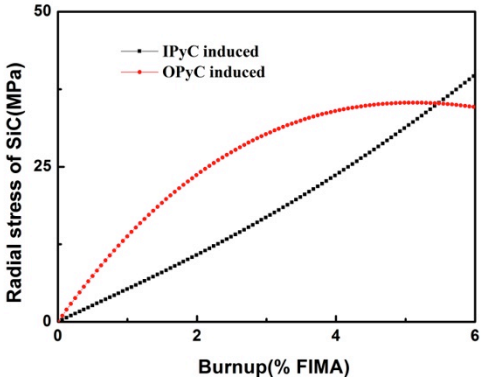

Fig. 3: Radial stress of SiC induced by irradiation swelling and creep of PyC.

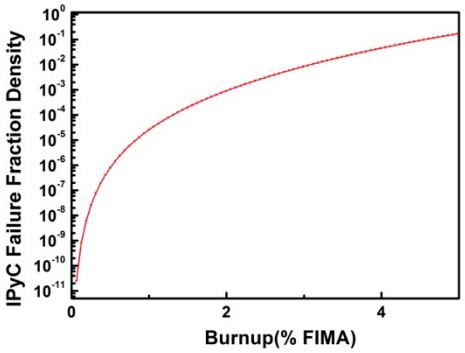

Fig. 4: Failure distribution density of IPyC

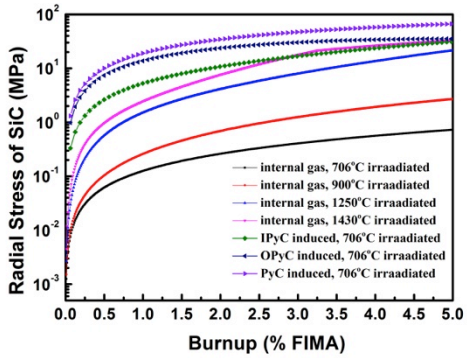

Fig. 5: Tensile and compressive stress in SiC

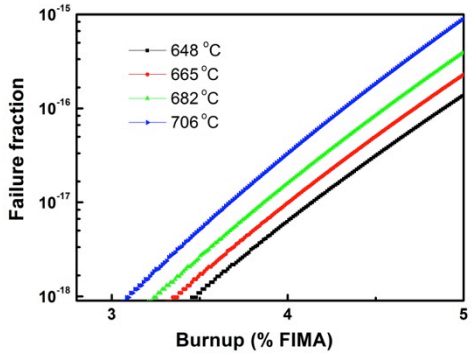

Fig. 6: Failure fraction under operating conditions during the designed lifetime of TMSR-SF.

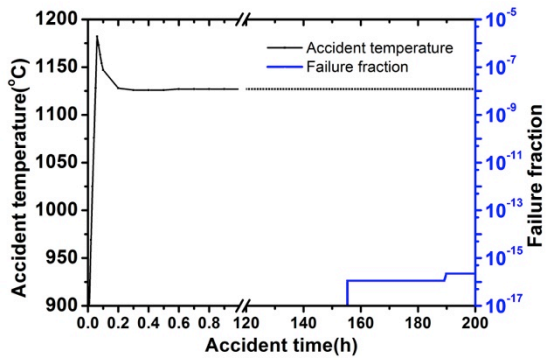

Fig. 7: Failure fraction of TMSR-SF under nominal accident conditions.

///. E. Fission Products Release Fraction

Adopted the diffusion coefficients as described in Ref.[8], the release fraction of radionuclides from kernel, particle and element for each species are calculated based on the method described above, as shown in Fig. 8. Besides the fractional release under operation condition, this figure also presents fractional release variation with time in the case of accident condition.

From Fig. 8, it is evident that the fractional releases of four radionuclides (^{137}Cs, ^{90}Sr, ^{110m}Ag, and ^{131}I) increase with the irradiation time. After about 250 EFPD, the fractional release of each species continuously increases with accident time. Fractional release curve verse time have the same trends except Iodine. Unlike other nuclides, release fraction of iodine almost keeps constant both in normal operation and accident condition. Anyway, during the nominal scenario, fractional releases of the four radionuclides keep extremely low level ($<10^{-5}$).

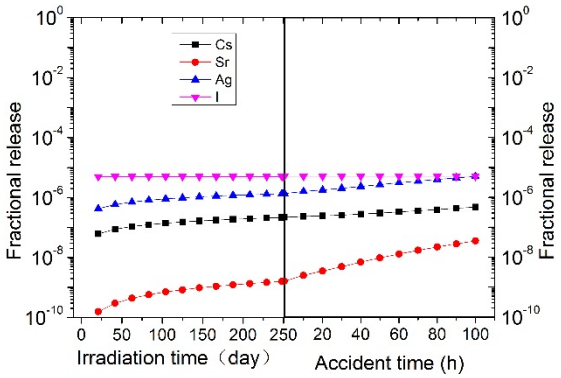

Fig. 8: Fractional release of ^{137}Cs, ^{90}Sr, ^{110m}Ag, and ^{131}I.

VI. Conclusion

On the basis of above calculation and analysis, the failure probability of TRISO fuel and fission products release fraction keep extremely low level (Fig. 6, 7, 8) under both normal operation and accident conditions in TMSR-SF. So the safety of TMSR-SF coated fuel should be guaranteed by using the advanced fabrication process. Therefore, the dominant contribution to the fuel failure fraction and fission products releasing is the free uranium and uranium contaminant introduced during the fabrication process.

Although, low failure fraction and fission products release fraction were expected, further studies should be performed on the compatibility between spherical fuel elements and molten salt. A series of verification tests are already in progress, and preliminary experiments are expected to be started at the end of 2014.

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REFERENCES

[1] The Aircraft Reactor Experiment, Nuclear Science and Engineering, 2, 797，1957.
[2] H. G. MacPherson The Molten Salt Reactor Adventure, NUCLEAR SCIENCE AND ENGINEERING: 90, 374-380 (1985)
[3] Bongartz K. Status of the Fuel Stress and Failure Rate Calculation at KFA: KFA Report. Juel-1686, 1980.
[4] Michael J Kania, Heinz Nabielek, Hubertus Nickel, Coated particles for the HTR, 2009
[5] Karl Verfondern, TRISO Fuel Performance Modeling and Simulation, Chapter 3.24 in ‘Comprehensive Nuclear Materials’ by Rudy Konings (ed.), Elsevier Ltd, 2012
[6] David Petti et al., “Development of Improved Models and Designs for Coated Particle Gas Reactor Fuels”, INEEL/EXT-05-02615, 2004
[7] IAEA Technical document, Advances in High Temperature Gas Cooled Reactor Fuel Technology, IAEA-TECDOC-1674, Austria, June, 2012
[8] IAEA Technical document, Fuel Performance and Fission Product Behaviour in Gas Cooled Reactors, IAEA-TECDOC-978, Austria, 1997.