Modeling of Coated Fuel Particles Irradiation Behavior

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Abstract: In this report, PANAMA code was used to estimate the CP performance under normal and accident condition. Under the normal irradiation test (1 000 ℃ 625 efpd, 10% FIMA), for intact CP fuel, failure fraction is in the level of 10^{-7}. As-fabricated SiC failed particles results in the through coatings failed particles much earlier than the intact particles does, OPyC layer does not fail immediately after irradiation starts. The significant failures start at beyond the burnup of about 7% FIMA.

Under the accident condition, the calculated results showed that when the heating temperature is much higher than 1 850 ℃, the failure fraction of coated particle can reach the level of 1 percent. The CP fuel fails significantly if it has a buffer layer thinner than 65 um, SiC layer thinner than 30 um. High burnup CP need to develop small size kernel, thick buffer layer and thick SiC layer.

Key words: HTGR, Coated particle, Modeling

1. Introduction

A characteristic feature of a High temperature gas-cooled reactor (HTGR) is the essential role of the TRISO coated fuel particle acting as a tiny containment and serving as the principal barrier against radionuclide release under normal operation and accident conditions. The quality of the fuel relies on minimizing the number of failures of these particles during reactor operation.

The 10 MW High Temperature Gas-cooled Reactor (HTR-10) built in China is a modular pebble-bed type HTGR with a thermal power of 10 MW. Spherical fuel elements are employed in the pebble-bed core. The HTR-10 fuel element with 60 mm in diameter consists of matrix graphitic material, in which TRISO coated fuel particles are dispersed. The coated particle is composed of a

central UO_2 kernel of 500 μm in diameter and four layers, which are (1) a low density porous buffer layer, (2) an inner high-density isotropic pyrolytic carbon (PyC) layer, (3) a silicon carbide (SiC) layer, and (4) an outer high-density isotropic PyC layer. About 20 000 spherical fuel elements were produced for the HTR-10 in 2000 and 2001. Each fuel element contained about 8 300 coated particles. The production batches showed a free U fraction of 1.4×10^{-4}; this corresponds to one to four defective particles in each irradiated fuel sphere.

The irradiation test of four HTR-10 spherical fuel elements was carried out in the IVV-2M research reactor at Zarechnyy, Russia. After the burnup of the irradiated fuel elements reached 100 000 MWd/t, a high temperature heating test with the fuel element in capsule 5 was performed in the reactor. Post-irradiation examination indicated that at normal irradiation condition, the PyC and SiC layers of particles kept their integrity. However, after the high temperature irradiation of the fuel element, the failure fraction of coated particles in this sphere reached 5.8%. The heating temperature was expected to be about 1 600 ℃, but the actual fuel temperature was presumably much higher than 1 600 ℃ because the thermocouple to measure the fuel element failed and it became impossible to record and to control the temperature exactly. In this report, the PANAMA fuel performance code was used to find out what the actual temperature during the heating test was by calculating the particle failure fraction under the test conditions and taking the temperature as a parameter.

2. Irradiation Samples and Irradiation Conditions

The fabrication technology of irradiation samples includes UO_2 kernel preparation by the gel precipitation method, PyC and SiC coating on the UO_2 kernels by the CVD process in a fluidized bed and manufacturing the spherical fuel element through a quasi-isostatic process. Tab. 1 shows the main characteristics of UO_2 kernel and coated particles.

In order to assess the performance of the fabricated fuel elements, four spherical fuel elements randomly sampled from the first production batch (1000 spherical fuel elements) and placed into separate irradiation capsules (No. 2 through 5), an in-pile irradiation test was carried out in the Russian IVV-2M research reactor. The irradiation rig contains 5 independent capsules, as shown in Fig. 1(a). The capsule 1 contained about 13 500 loose coated fuel particles

and 60 samples of graphite matrix. The four spherical fuel elements were located in capsule 2 to capsule 5, respectively. Each capsule was independently controlled and continuously swept and monitored for volatile fission product release. The irradiation temperature was adjusted by He/Ne mixture gas.

Tab. 1 Main characteristics of UO_2 kernel and particle coating of HTR-10 first loading fuel

UO_2 diameter/μm	498
UO_2 density/(g/cm^3)	10.9
O/U ratio	2.00
Buffer PyC layer thickness/μm	95
I-PyC layer thickness/μm	42
SiC layer thickness/μm	37
O-PyC layer thickness/μm	42
Buffer PyC layer density /(g/cm^3)	0.98
I-PyC layer density /(g/cm^3)	1.86
SiC layer density /(g/cm^3)	3.20
O-PyC layer density /(g/cm^3)	1.87
I-PyC and O-PyC layer OPTAF	1.03

The nominal irradiation temperature was 1 000 ℃ ± 50 ℃. Their fast neutron fluence for this four capsules reached 1.10×10^{21}, 1.31×10^{21}, 1.30×10^{21} and 1.06×10^{21} n/cm^2, respectively. During the irradiation test, the temperature of the fuel element in capsule 3 was increased to 1 200 ℃ for 200 hours and to 1 250 ℃ for 200 hours, when its burnup reached 38 700 MWd/t and 57 000 MWd/t, respectively.

For the fuel element in capsule 5, the irradiation time was 625 efpd and the irradiation temperature was 1 000 ℃ ± 50 ℃. At the end of the normal irradiation test, the high temperature heating test was carried out in the reactor for 22 hours by increasing the fast neutron fluence and adjusting the ratio of He/Ne mixture gas. The post irradiation examination program included disintegration of the irradiated spheres and determination of the distribution of solid fission products in the matrix material along the sphere diameter, measurement of the failure fraction of the loose coated fuel particles obtained

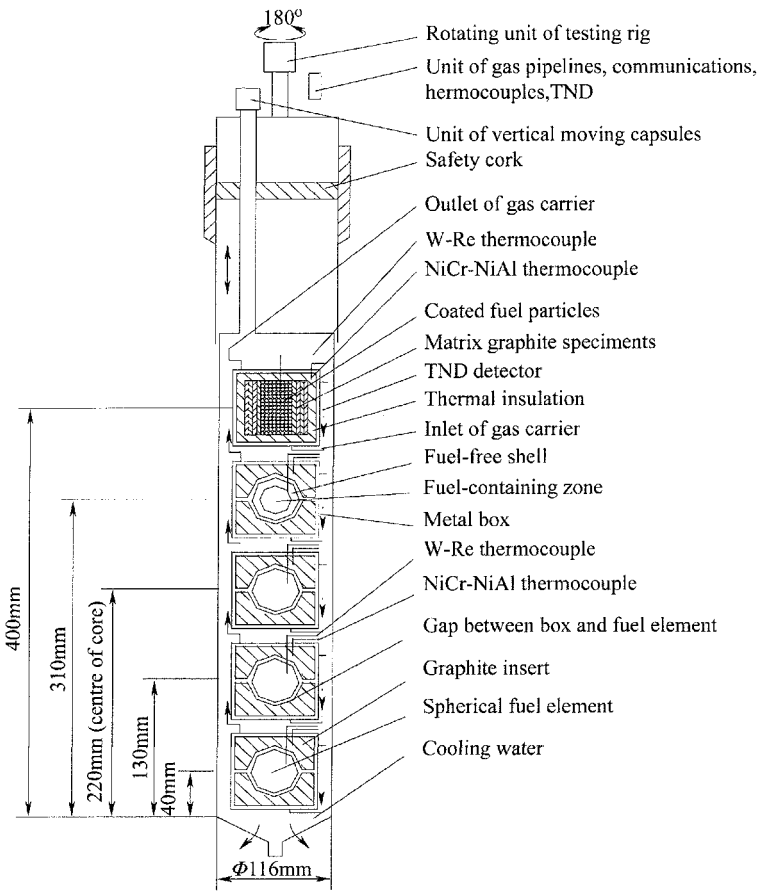


Fig. 1(a) Irradiation test rig for HTR-10 fuel elements

from the ball disintegration by the Irradiated Microsphere Gamma Analyzer (IMGA), and examination of the loose particles by ceramography.

The measurement of R/B of $^{85}Kr^{m}$ of the irradiated fuel elements in these four capsules as a function of the burnup is shown in Fig. 1(b). The free U levels of the fuel elements in the first production batch was 1.4×10^{-4}, it means that there might be 1～4 defect coated fuel particles in each irradiated fuel element due to the manufacture. After irradiation, the R/B of $^{85}Kr^{m}$ of fuel elements in capsule 2 and capsule 5 are in the range of 5×10^{-6} to 8×10^{-5}, the comparatively high R/B level was resulted from the higher manufacturing defects. All the release rate curves kept small fluctuation under average fuel

temperature, ~1 000 ℃. A little ascension of the curves at a point of about 53 000 MWd/tU came most likely from the fluctuation of the irradiation temperature.

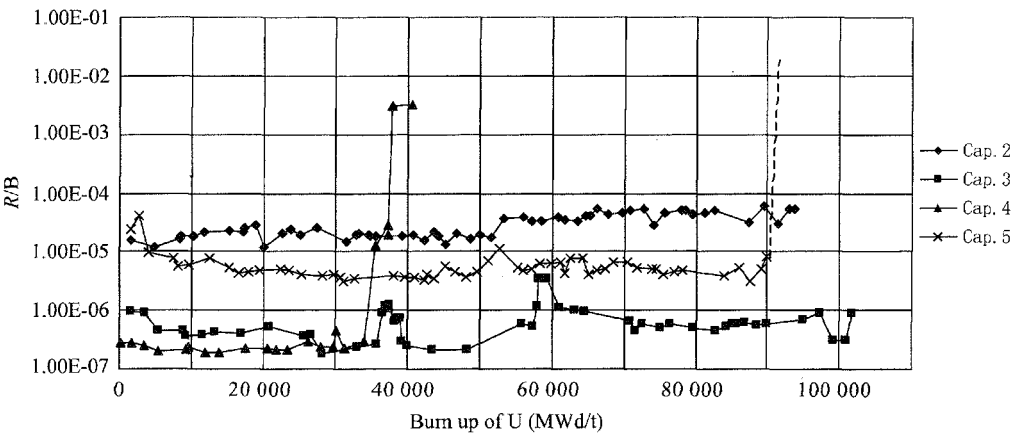


Fig. 1(b) R/B of $^{85}Kr^{m}$ of the irradiated fuel elements as a function of the burnup (the dotted line represents the R/B of $^{85}Kr^{m}$ for heating test)

Heating the fuel element in the capsule 3 to 1 200 ℃ for 200 hours with the burn up of 38 700 MWd/t, and to 1 250 ℃ for 200 hours with the burn up of 57 300 MWd/t caused an increase of about one order of $^{85}Kr^{m}$ release. When the temperature returned to 1 050 ℃, the release rate was restored to the initial value.

The irradiating testing of one fuel element in capsule 4 ended up early owing to something wrong in the gas loop of the capsule after irradiated time reached 5 349 effective hours (fuel burnup: 37 000 MWd/t).

3. CP Fuel behavior under irradiation

3.1 UO_2 kernel

Point defects, gaseous and solid fission products and oxygen are formed in kernel under irradiation, gas pebbles in fuel lead to the swelling of kernel and reduce the thermal conductivity of kernel. Experimental measurement suggested that larger values of 0.6% ~ 1.5% $\Delta V/V$ produced for per atom percent burnup, at 10% FIMA, 6%~15% increase in volume of kernel will be happen. Large amount of swelling can reduce void volume in CP, under some case cause

kernel/coating mechanical interaction.

The yield of Xe, Kr gas is about 0.31 per fission.

Free oxygen can form CO gas in CP fuel. For per fission, two oxygen atoms are released, about 1.4 atoms are tied up by rare earth fission products, other oxygen react with carbon to produce CO. Under irradiation:

$$\log(O/F) = -10.08 - 8\,500/T_i + 2\log t_i \quad (1)$$

Where T_i is the irradiation temperature (k), ti is irradiation time (s)

During heating:

$$\log(O/F) = -10.8 - 8\,500/T_i + 2\log T_i - 0.404\,(10\,000/T_h - 1\,000/(T_i + 75)) \quad (2)$$

Where Th is heating temperature (k)

Some fission products, especially Pd can migrate into contact with SiC layer, causing it to corrode. From experiment data, Pd corrosion rate (μm/h):

$$S = 2.613 \times 105\exp(-2.522 \times 105/8.314T) \quad (3)$$

Where T is the irradiation temperature (k).

3.2 Buffer layer

Under irradiation, Buffer layer will be densification rapidly (less than 4% FIMA), then it will be fracture. The densification of buffer layer can result in the increase of void volume of CP.

3.3 IPyC and OPyC layer

PyC is a brittle material, under irradiation and temperature, its anisotropy factor (BAF) and volume will be changed. Creep will happen in this layer, creep can relax its internal stress. By the act of neutron, PyC layer will undergo shrinkage as a result, tensile stress formed. This shrinkage can produce compress stress in SiC layer which is useful to reduce the tensile stress in SiC.

If a crack produced on the interface of IPyC/SiC, stress concentration will exist at crack tip on SiC internal face. Particle with the cracked IPyC has a SiC layer in tension state.

3.4 SiC layer

SiC has a high resistance against irradiation, it is considered to be the candidate for the first wall material in fusion reactor. At high temperature (above 1 600 ℃), thermal decomposition of SiC will occur. The decomposition temperature in different pressure, gas composition, should be calculated by HSC thermodynamic software.

4. Failure mechanisms of CP fuel

CP fuel failure comes from manufacture and operating in reactor. Now manufacture failure fraction is less than 10^{-5}, deformed particles and missing coating layers are now rare, damage to particles during pebble formation is rare, in-service failures received major interest. In order to increasing burnup and neutron fluence, set up VHTR (very high temperature gas cooled reactor), it is necessary to understand failure mechanisms.

Usually, in-service failure includes: pressure vessel (PV), SiC failure, OPyC shrinkage failure, and, interaction of CP and graphite matrix.

PV failure means if gas pressure exceeds coating layer UTS, the layer fail.

$$\sigma = rP/2t \tag{4}$$

Here, r is radius of the layer, P, gas pressure, t, layer thickness.

$$P = P_{Kr} + P_{Xe} + P_{CO} \tag{5}$$

SiC failure includes: kernel migration (Amobe effect); FP corrosion; cracks in SiC layer; thermal decomposition; U dispersion.

Crack in SiC layer is mainly caused by IPyC crack, because in this case, stress concentration will exist at crack tip on SiC internal face, as shown in Fig. 2, stress concentration speed up the growth rate of flaws. Fig. 3 is a cracked SiC layer, this failure is correlated with IPyC crack.

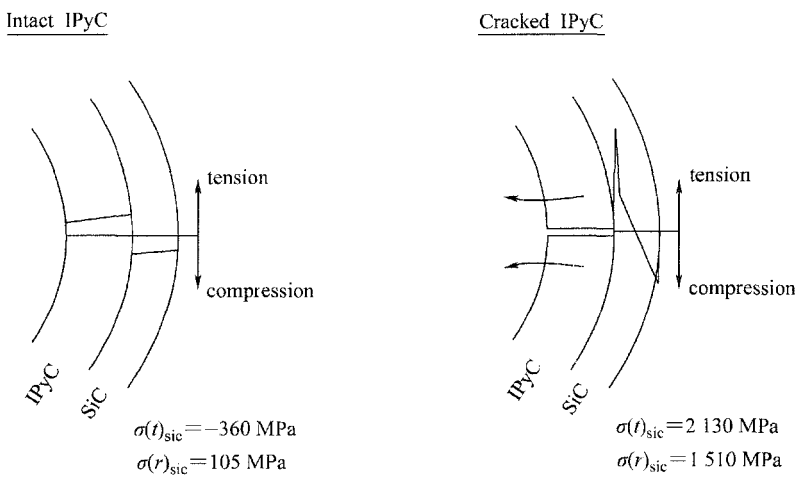


Fig. 2 stress in SiC layer

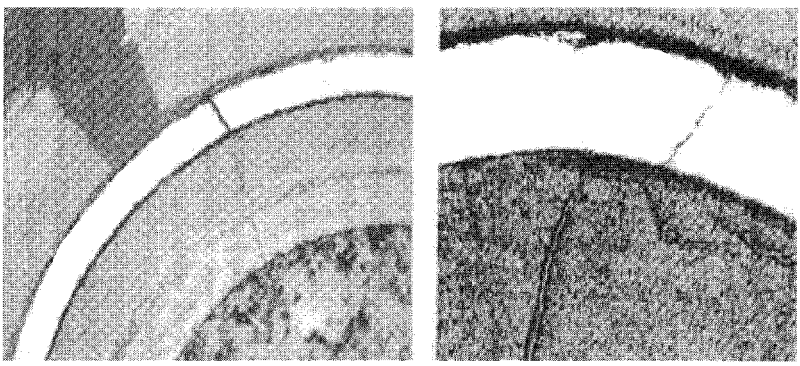


Fig. 3 Cracks in SiC layer

5. PANAMA code

PANAMA code of CP during irradiation and under accident condition (i. e., heating) with temperature up to 2 000 ℃ is based on the pressure vessel model, where SiC layer represents the actual retention barrier for fission products. In this code, a particle fails when stress in the SiC layer induced by the internal gas pressure exceeds the SiC strength.

Failure fraction is given by

$$\Phi_{\text{total}} = 1 - (1 - \Phi_0)(1 - \Phi_1)(1 - \Phi_2) \tag{6}$$

Where Φ_1 and Φ_2 correspond to PV failure fraction and thermal decomposition failure fraction, respectively. Φ_0 is the manufacture failure fraction, at this time, is less than 10^{-5}, so in this code $\Phi_0 = 0$ is reasonable.

If no heating test after irradiation, or at the beginning of heating, time $t_h = 0$, $\Phi_2 = 0$.

At time t after the start of heating test at temperature T, PV failure fraction is given by

$$\Phi_1(t, T) = 1 - \exp[-\ln 2(\sigma_t / (\sigma_0)^m] \tag{7}$$

σ_t represents stress at t time due to the gas pressure, σ_0 is strength at the end of irradiation, m is weibull parameter. Strength values (pa) are scattered in a Weibull distribution.

$$\sigma_t = 0.5rP/d_0(1 + ct/d_0) \tag{8}$$

Where r is average radius of SiC layer, P is fission gas pressure, c is corrosion rate as a function of temperature, d_0 is original thickness of SiC layer.

$$P = BRT(0.31F_d + OPF)/(V_m V_f/V_k) \quad (9)$$

Where B is burnup, R is gas constant, F_d is relative fraction of fission gas releases, OPF is number of oxygen atoms per fission, V_m is molar volume of kernel, V_k is kernel volume, V_f void fraction, 50% for buffer layer.

After irradiation, if CP is heated at 1 600~2 500 ℃, particle will fail due to the thermal decomposition of SiC layer, failure fraction is

$$\Phi_2(t,T) = 1 - \exp(-\xi\alpha^\beta) \quad (10)$$

Where α、β are empirical constant, ξ is the action integral as a function of temperature. In PANAMA code, α is 1×10^{-4}, and β is 4.

6. Modeling of CP irradiation behavior

Tab. 2 shows the geometry and key specification of the CP used in this report. Irradiation condition: The maximum burn up reached 10% FIMA, the maximum fast neutron fluencies is 1.06×10^{25} n/m^2. Irradiation temperature is (1 000±50)℃, irradiation time is 625 efpd.

Tab. 2 The geometry and specification of CP

UO_2 kernel	
Diameter (μm)	498
Density (g/cm^3)	10.8
O/U	2.0
CP	
Buffer layer thickness (μm)	95
IPyC layer thickness (μm)	45
SiC layer thickness (μm)	37
OPyC layer thickness (μm)	42
Buffer layer density (g/cm^3)	1.1
IPyC and OPyC density (g/cm^3)	1.8
SiC density (g/cm^3)	3.18
OPTAF of PyC	1.03

6.1 Boundary conditions for calculations

One of the most important input parameters for PANAMA code is SiC strength and its weibull parameter. In this report we used several data of SiC

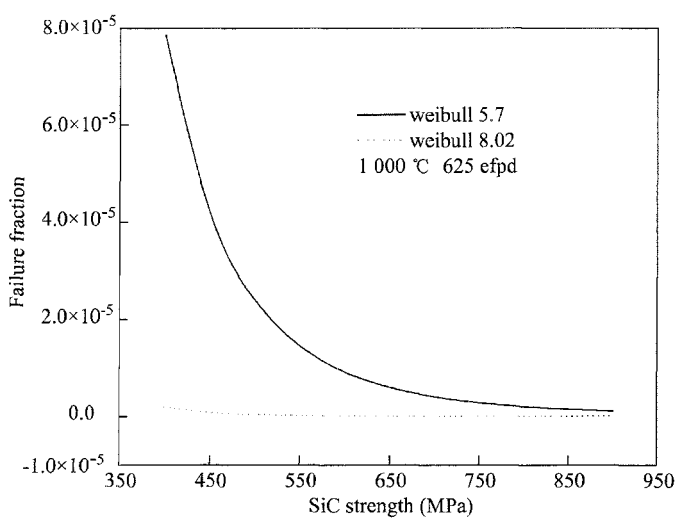
strength and weibull parameter, and changed some CP geometry and heating temperature in order to get more knowledge about PANAMA code and CP irradiation behavior.

(1) Failure fraction under normal irradiation

CP undergoes normal irradiation, i. e. , irradiation temperature is 1 000 ℃, irradiation time is 625 efpd. Tab. 3 and Fig. 4 show the failure fraction.

Tab. 3 Failure fraction and SiC boundary conditions(10% FIMA)

SiC strength (MPa)	Weibull parameter	Failure fraction
834	8.02	8.09E−9
700	8.02	3.21E−8
600	8.02	9.43E−8
400	8.02	1.94E−6
300	8.02	1.66E−5
834	5.7	1.60E−6
700	5.7	5.63E−6
600	5.7	9.14E−6
400	5.7	1.54E−5
300	5.7	3.60E−4



(a)

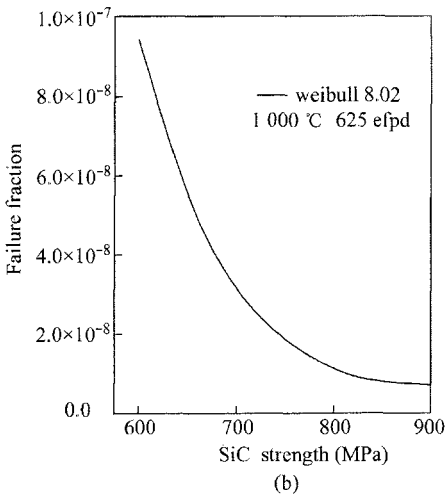


Fig. 4 Failure fraction as a function of SiC strength

It can be seen that failure fraction is more sensitive to weibull parameter, under normal irradiation condition, a CP with an intact SiC layer, even if the SiC layer has a low strength (300 MPa) and a bad strength distribution (weibull parameter is 5.7), the failure fraction is still less than 5×10^{-4}, because the assumption of PANAMA code is somewhat conservative, we can conclude that for a SiC intactness CP, at normal operating condition, failure fraction can meet designed safety requirement.

In fact, SiC strength value exceeds 700 MPa and weibull larger than 5.7 can be trusted by the present manufacture technology. Calculated stresses on each layer of a intact particle by K. Sawa showed that the maximum tensile stress on the SiC layer is almost zero when the burnup near 5% FIMA, when irradiation starts, compressive stress acts on the SiC layer by fast neutron-induced shrinkage of the PyC layers. PANAMA code does not consider the act of PyC layers, this is one reason resulted the conversitivisms of this code. Under this normal irradiation test, failure fraction is in the level of 10^{-6} is reasonable.

In the case of CP with as-fabricated SiC defects, or in the case of SiC failed, PANAMA code assumed that the CP failed as soon as SiC failed. Here we considered that OPyC layer is still intact, it served pressure vessel wall against internal gas pressure, Fig. 5 shows the failure fraction of a CP with failed SiC

layer. PyC layer strength is 160 MPa, weibull parameter is 4.

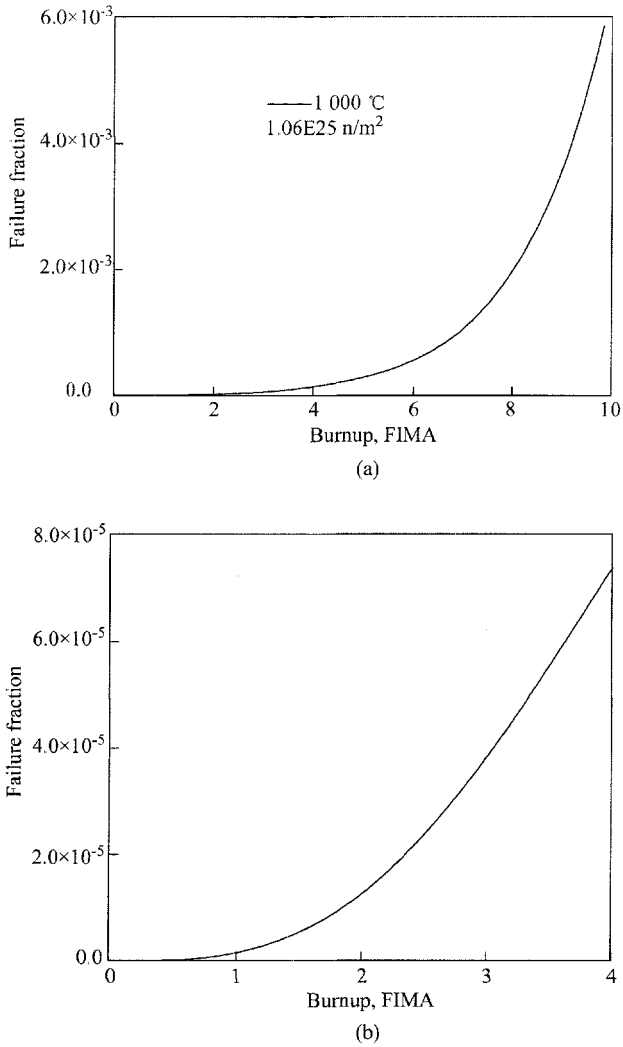


Fig. 5 Failure fraction for OPyC layer as PV wall under normal irradiation

Compared Fig. 4 with tab. 2, it is indicated that the as-fabricated SiC failed particles result in the through coatings failed particles much earlier than the intact particles does, OPyC layer does not fail immediately after irradiation starts. The significant failures start at beyond the burnup of about 7% FIMA.

(2) Failure fraction under accident condition

If SiC strength is 834 MPa, weibull parameter is 8.02, after irradiation at 1 000 ℃ for 625 efpd, then heating the irradiated CP fuel at 1 570 ℃ for 200 hours, Fig. 6 shows the relationship between failure fraction and heating time. At the end of 200 hours, failure fraction remains below 5.3×10^{-4}.

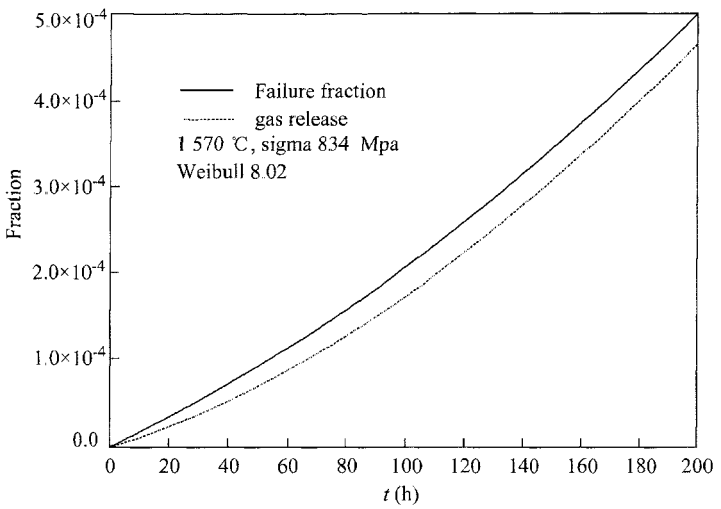


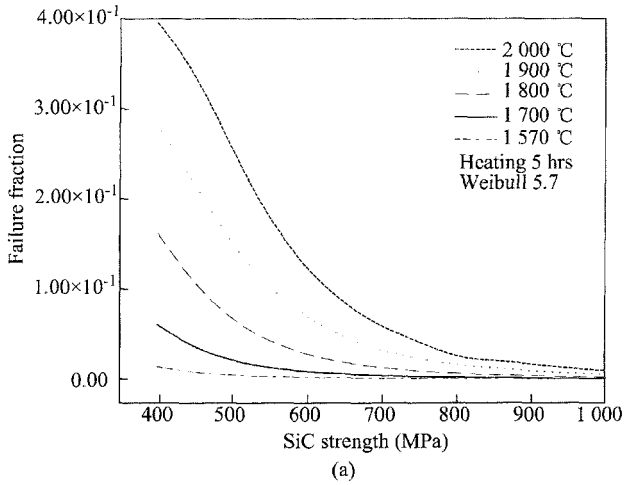
Fig. 6 The relationship between failure fraction and heating time

In this report we made an assumption: SiC strength is not 834 MPa, and the strength distribution is not good, here we regarded it as 5.7. After irradiation at 1 000 ℃ for 625 efpd, burnup reached 10% FIMA, CP fuel underwent heating test from 1 570 ℃ to 2 000 ℃ and keep CP in this temperature for 5 hours. The total heating (annealing) time is 22 hours. Fig. 7 shows the calculation results.

From Fig. 7, we chose some suitable SiC strength and heating temperature, make the CP failure fraction fall in 10^{-2} level.

It can be seen from Fig. 8, if the heating temperature is 1 570 ℃, and the SiC strength less than 425 MPa, failure fraction will reach 10^{-2}. When the heating temperature higher than 1 900 ℃, even the SiC has a high quality (strength is 834 MPa), failure fraction is still reached 10^{-2}.

Normally, the accidental temperature in HTR is below 1 600 ℃, in this condition, when the SiC strength less than 425 MPa, failure fraction will be 10^{-2}.



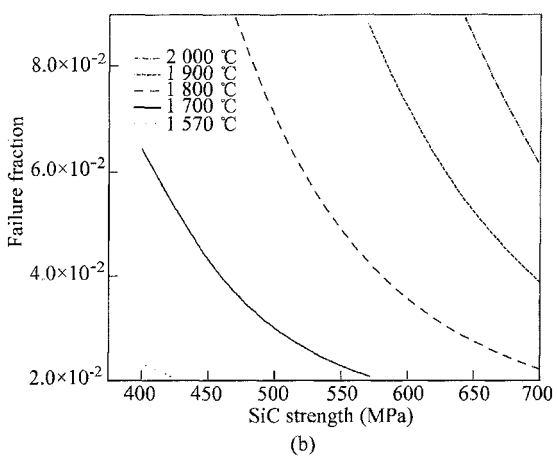


Fig. 7 Failure fraction for different SiC strength heating at some temperatures

The probability is: 420 MPa/1 570 ℃, 570 MPa/1 700 ℃, 700 MPa/1 800 ℃. Fig. 8 is the some temperatures and values of SiC strength which 10^{-2} failure fraction can be obtained. Large experimental data indicated that when the heating temperature above 1 800 ℃, PANAMA code showed good agreement with the CP fuel irradiation data. Below 1 800 ℃, the attribution of SiC thermal decomposition to CP fuel failure is not so large like the predication by PANAMA code, and annealing can relax tensile

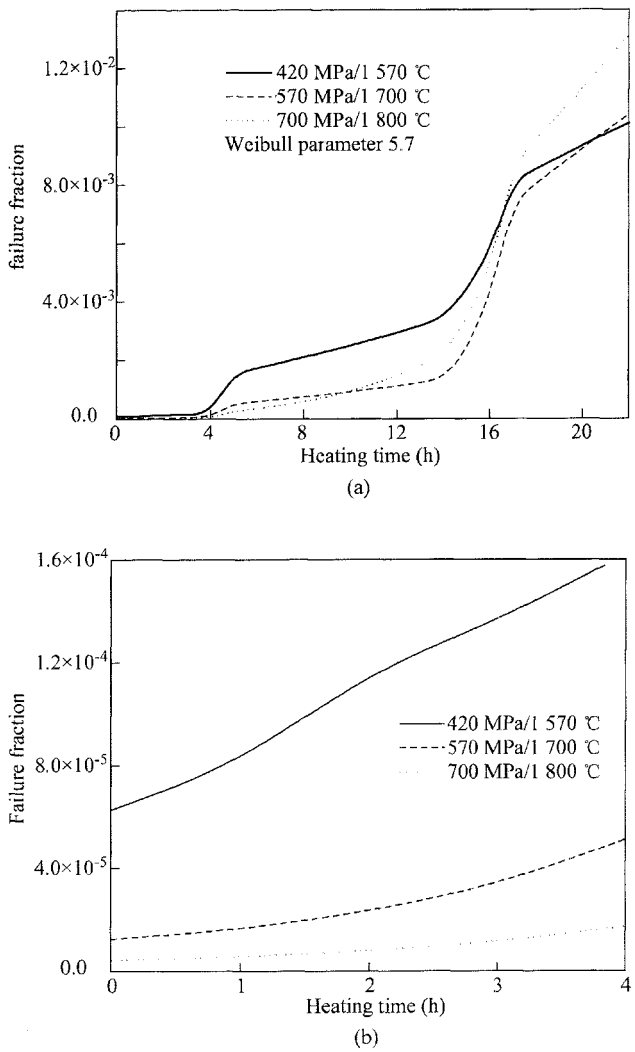


Fig. 8 possibility of failure fraction reaches 10^{-2}

stress in the SiC layer, this is another reason resulted the conversativisms of PANAMA code. On the other hand, SiC strength is at least 700 MPa, so we can conclude that for an intact CP, only when the heating temperature is 1 800 ℃ or above 1 800 ℃, failure fraction can be larger than 1×10^{-2}.

If CP exist as-fabricated SiC defects, through-coating failure fraction is decided by the strength of OPyC layer. Fig. 9 shows the failure fraction changes

as the heating time. At the end of heating test, failure fraction is 1.34×10^{-1}.

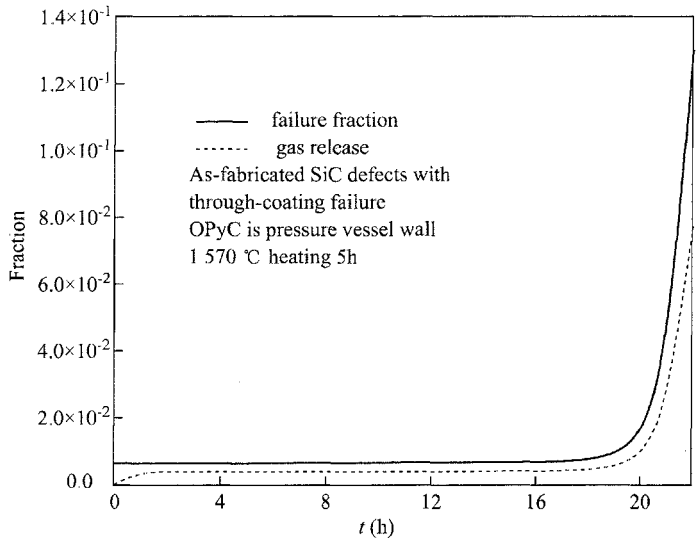


Fig. 9 Failure fraction as a function of heating time for as-failed SiC CP

(3) Effect of CP geometry on failure fraction

Boundary conditions:

Irradiation time: 625 efpd

Irradiation temperature: 1 000 ℃

Fast neutron fluence: 2.1×10^{25} n/m^2.

Burnup: 10% FIMA

Heating temperature: 1 600 ℃

Heating time: 200 h

SiC strength: 834 MPa

Weibull parameter: 8.02

Fig. 10 shows the relationship between CP geometry and failure fraction. Because PANAMA code did not consider the effect of OPyC layer, the changing of the thickness of OPyC layer, failure fraction has no change.

The irradiation behavior of IPyC layer is not be treated in PANAMA code, but the radius of SiC layer decreases with the decreasing of IPyC thickness, tensile stress in SiC layer is written as $rP/(2t)$, if r decreases, t increase, tensile stress will be reduced, as a result, failure fraction reduces. So the failure

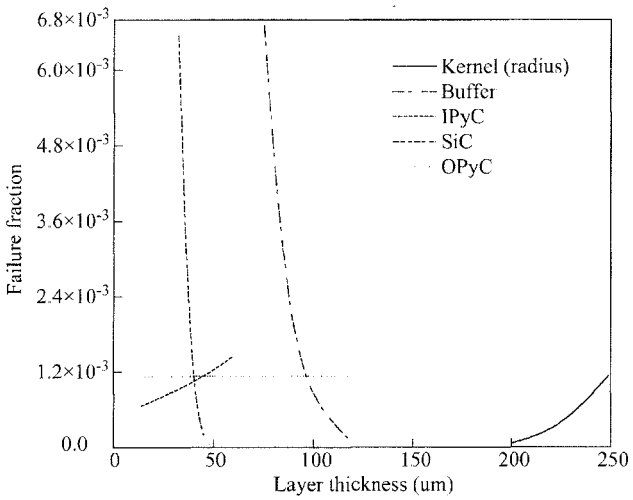


Fig. 10 The relationship between CP geometry and failure fraction

fraction decreases by the decreasing of IPyC thickness or increasing of SiC thickness.

Fig. 10 indicated that there are two key parameters affect failure fraction, one is SiC layer thickness, the other is buffer layer thickness. The calculated results predicted that at accident condition, the CP fails significantly if it has a buffer layer thinner than 65 μm, SiC layer thinner than 30 μm. High burnup CP need to develop small size kernel, thick buffer layer and thick SiC layer.

7. Summary

The PANAMA fuel performance code was used to estimate the CP performance. Under the normal irradiation test (1 000 ℃ 625 efpd, 10% FIMA), for intact CP fuel, failure fraction is in the level of 10^{-7}. As-fabricated SiC failed particles results in the through coatings failed particles much earlier than the intact particles does, OPyC layer does not fail immediately after irradiation starts. The significant failures start at beyond the burnup of about 7% FIMA. At accident condition, 5 hours, for an intact CP, only when the heating temperature is 1 800 ℃ or above 1 800 ℃, failure fraction can be larger than 1×10^{-2}; If CP exist as-fabricated SiC defects, through-coating failure fraction is 1.34×10^{-1}.

Under heatup accident condition, the CP fuel fails significantly if it has a buffer layer thinner than 65 μm, SiC layer thinner than 30 um. High burnup CP need to develop small size kernel, thick buffer layer and thick SiC layer.

PANAMA code is a very simple but useful tool for prediction CP fuel irradiation, we can use it to design CP geometry, plan irradiation test, and guide us to observe CP during PIE.

Note

This work was performed by Dr. Tongxiang Liang during his visit in CEA (Cadarache) from November,2003 to October,2004

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