A Universal Model for Coated Particle Failure

K. Bongartz, H. Nabielek
Kernforschungsanlage Jülich GmbH,
Postfach 1913, D-5170 Jülich 1, Germany

Summary

The design of the helium-cooled High Temperature Reactor (HTR) is unique in that a single core may contain from 2 to 30 billion miniature "fuel elements" in the form of small coated spherical kernels. Each of these coated particles has an outer diameter of less than one millimetre. For easy core management, thousands of coated particles are embedded in a graphite matrix structure to form larger individual fuel units such as the 60 mm spherical (THTR) or 360 x 790 mm hexagonal block type (Fort Saint Vrain) HTR fuel elements now in use.

The particle coatings serve as pressure vessel for the fission gases and primary diffusion barrier to solid fission product release and must retain their integrity for 3 - 4 years at temperatures around 1000 ^{o}C. The mechanisms of in-pile failure for such a fuel kernel coated with a silicon carbide (SiC) layer prestressed by inner and outer pyrocarbon (PyC) layers are well known. The statistical occurence of pressure vessel failure could be successfully predicted with the so-called STRESS code from the measured statistical distributions of particle parameters such as kernel diameter and coating thickness and the distribution of the SiC layer strength. From experimental work both in and out-of-pile, models and data are being developed which describe the progression of kernel migration into the PyC (amoeba-effect), SiC attack and SiC decomposition. The onset of failure due to amoeba effect is assumed when the fuel kernel reaches the inner SiC surface. SiC corrosion is modeled assuming progressive thinning due to fission product attack. Failure is marked by the breach of the SiC coating serving as pressure vessel. Onset of failure is accelerated by thinning of the layer as well as by reduction of its specific strength.

In a realistic irradiation environment all three mechanisms will show mutual interaction. An attempt has been made to create a Universal Particle Model which includes all these mechanisms and also their interactions.

1. Introduction

Failure mechanisms of coated particles with a SiC interlayer occuring during irradiation at high burn-ups of 100 000 Md/t and at temperatures of 1000 - 1500 ^{o}C are well known:

- Build-up of internal gas pressure leading to pressure vessel failure
- Amoeba effect, i.e. fuel migration in thermal gradients results in an attack of the coating
- SiC corrosion reducing thickness, strength and impermeability of this most important layer.

The STRESS code for the prediction of particle failure up to now took into account pressure vessel failure alone. In the test BR2-P21 all failure mechanisms mentioned above were observed to a large extent in post irradiation examination. It was therefore ideally suited to serve as a guide for the extension of the STRESS model taking into account the simultaneous influence of amoeba attack and SiC corrosion as an accelerating factor for pressure vessel failure.

2. Recent modifications

The development of models predicting particle failure starting with the thin shell approach via the Walther model /1/ to the present version enabling predictions of the 10^{-6} failure level is well documented /2/ /3/. Recent essential modifications were the use of a more realistic description of the Xe, Kr /4/ and CO release /5/ /6/. In the STRESS code kernel swelling also can fail particles independent of gas pressure build-up. Under the conditions of the BR2-P21 experiment the rather unusual event of particle failure due to transmission of solid state pressures from the swelling kernel which comes into contact with the coating is predicted. The calculated steep increase in fig. 1 clearly overestimates observed failure rates. Observations on irradiated particles show amoeba attack and SiC corrosion as dominant failure mechanisms /7/. Therefore, the mechanical interaction between kernel and coating has been removed from the STRESS code. The lower curve in fig. 1 will be used as a basis to study chemical effects on pressure vessel failure.

3. Chemical Effects

Chemical effects contributing to coated particle failure are amoeba attack and SiC corrosion.

The amoeba attack involves heavy metal migration towards the hot end of the particle whereby carbon from the PyC layers is removed and deposited at the cold end and for the UO_2 fuels the rate law is given by /8/ and for (Th, U)O_2 by /9/.

In order to simulate amoeba attack failure has been predicted by varying the thickness of the inner PyC layer for the particles irradiated in BR2-P21. Fig. 2 shows the small effect in failure prediction even with only 1 /um of inner PyC left. Further work in dynamic representation of amoeba attack rate has therefore been abandoned.

Silicon carbide corrosion can effect coated particle failure in several ways

- Reduction of SiC strength due to bulk corrosion, this was simulated in the calculated failure rates in fig. 3 by a variation of the strength of the SiC.
- Broadening of the SiC strength distribution (a Weibull distribution is assumed) due to grain boundary corrosion and creation of large microcracks. This was simulated in the calculated curves in fig. 4 by a variation of the Weibull parameter.
- Thinning of the layer thickness by assuming a corrosion zone without any strength, simulated in the calculated curves in fig. 5 by a variation of the SiC thickness.

The first attempts in simulating these effects with the STRESS code were performed by a variation of strength, Weibull parameter (i.e. width of the strength distribution) and SiC layer thickness. Figures 3 through 5 show that strength reduction and SiC thinning are effective in reproducing observed failure levels. For practical reasons - SiC thinning is investigated in laboratory and irradiation experiments - we decided to build a dynamical representation with SiC corrosion depth rate law equ. 2 into the code. The limiting hoop stress failure criterion is still retained. Failure probability, however, is enhanced by the decreasing thickness of the SiC layer. It is obvious that SiC corrosion is one reason for particle failure in the BR2-P21 experiment fig. 6 /7/.

If we assume a reaction rate limited corrosion mechanism we expect SiC thinning y to behave according to a law like

$$y = Kt^n \text{ with } K = K_0 \exp(-Q/RT) \qquad (1)$$

We have determined the constants in equ. (1) by comparison between observed failure levels and the prediction for 17, 26 and 31 /um SiC thinning (fig. 5) in BR2-P21 to give the rate law

$$\log y = -4.70 - 0.65 \times 10^4/T + 0.5 \log t \qquad (2)$$

with y (m), T irradiation temperature (^{o}C) and t irradiation time (s). Surprisingly, both reaction order (n = 0.5) and activation energy (Q = 124 kJ x mol^{-1}) are quite similar to values observed with carbide fuels in laboratory experiments involving a temperature gradient of 300 $^{o}C\ cm^{-1}$ /10/.

4. The BR21-P21 Experiment

This experiment was a fuel performance test performed at 1350 and 1550 ^{o}C respectively up to a burn-up of 11 % fima duration 380 days. Post irradiation examination of coated particles revealed massive SiC corrosion, amoeba attack and pressure vessel failure. The essential components for the successful description of particle failure in BR21-P21 are

- STRESS model predicting statistics of failure by combining the normal distribution in fuel kernel diameter and coating layer thickness with the strength distribution of the SiC layer according to Weibull,
- no mechanical interaction between fuel kernel and coating in high fluence experiments,
- reduction of SiC thickness according to equ. (2) i.e. 31 $/_{um}$ at 1550 ^{o}C and 11 $/_{um}$ at 1350 ^{o}C after 350 days of irradiation.

Fig. 7 shows the comparison between observed and calculated particle failure in the 1550 ^{o}C capsule of BR2-P21. Up to 3 % fima the gas release is determined by heavy metal contamination and particle breakage during fuel fabrication. At higher burn-ups the agreement between model and observation is very good. The onset of particle failure at the 10^{-5} to 10^{-4} level is described correctly and the evolution of failure during irradiation is being followed with high precision. This result gives high confidence both in the mathematical model employed and in the proposed mechanisms of SiC thinning.

For the other capsule of the experiment run at 1350 ^{o}C the onset of in-pile particle failure is equally described with high precision at 6 % fima (fig. 12). Towards the end of irradiation the observed failure levels increase by a factor 10 beyond predicted levels. At this point all thermocouples have failed and temperatures have been estimated at 1400 ^{o}C. We assume that the reason for the discrepancy is a unaccounted temperature excursion to temperatures above 1400 ^{o}C.

5. Conclusion

The STRESS code in its present form is the product of continuous development over a decade. Rather than developing new models, the basic structure of mechanisms has been retained to accomodate evolutionary improvements towards a Universal Model for coated particle failure. One large step forward has been fast execution times to enable Monte Carlo predictions at failure incidence at 10^{-4} level. Now it is important that the code is flexible enough to include all observed failure mechanisms. The success in the case of one particularly intricate irradiation experiment with massive silicon carbide corrosion was encouraging, because both incidence and progression of failure were predicted precisely.

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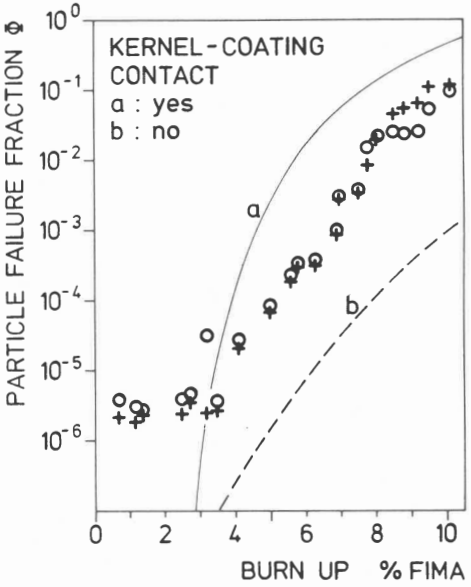

Fig. 1: Predicted and observed failure fraction in BR2-P21/2. The failure mechanism whereby solid state forces originating in kernel swelling can fail Triso particles is taken into account (curve a) is neglected (curve b) in the calculation.

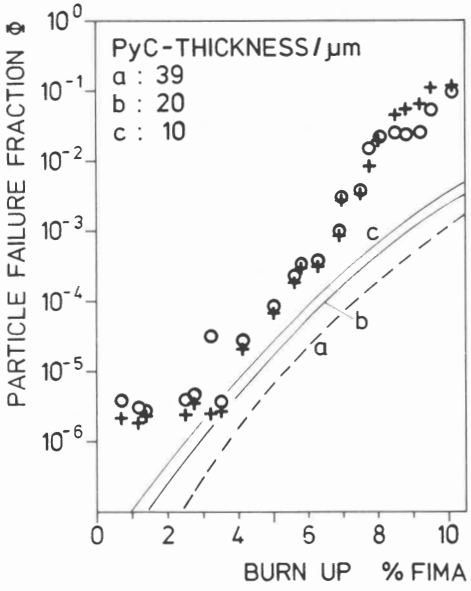

Fig. 2: Predicted and observed failure fraction in BR2-P21/2. Amoeba attack is simulated by a variation of the PyC layer thickness in the calculated curves.

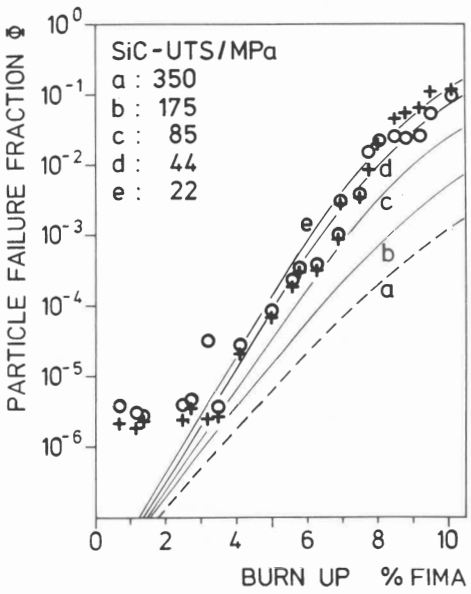

Fig. 3: Predicted and observed failure fraction in BR2-P21/2. The ultimate tensile strength (UTS) of the SiC layer is varied in the calculated curves.

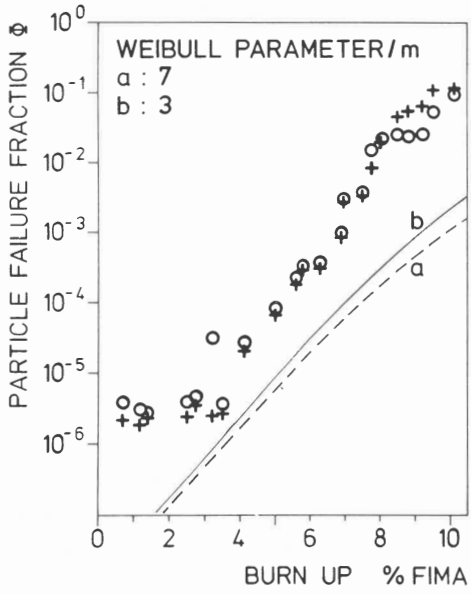

Fig. 4: Predicted and observed failure fraction in BR2-P21/2. A potential effect of SiC grain boundary corrosion in widening the strength distribution is simulated by a variation of the Weibull Parameter m in the calculated curves.

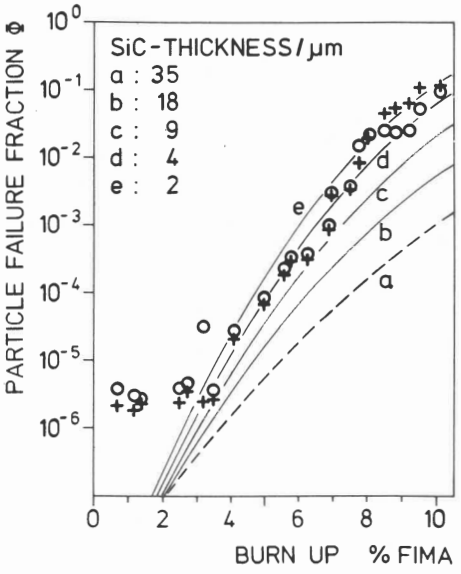

Fig. 5: Predicted and observed failure fraction in BR2-P21/2. SiC-corrosion is simulated by a variation of the SiC thickness in the calculated curves.

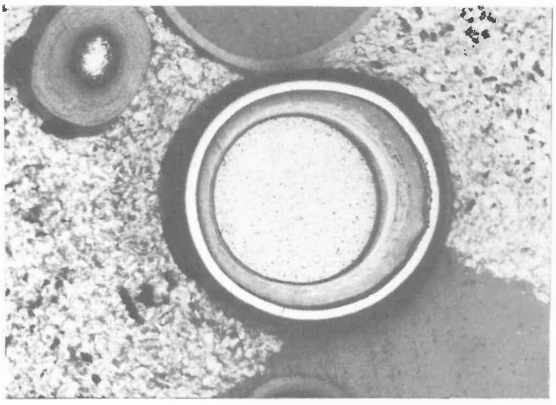

Fig. 6: SiC corrosion at cold end and amoeba attack at hot end of one particle in BR2-P21.

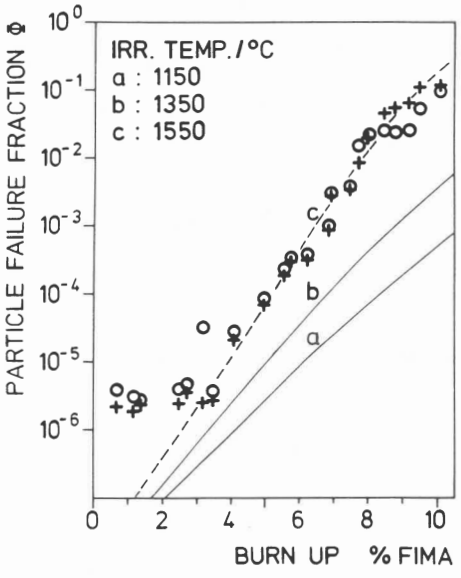

Fig. 7: Observed failure fraction in BR2-P21/2 (irradiation temperature = 1550 °C) in comparison to modified STRESS code prediction with corrosion law equ.(2)

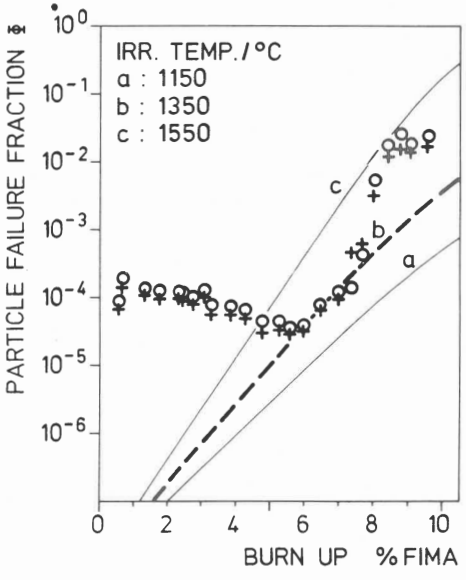

Fig. 8: Observed failure fraction in BR2-P21/1 (irradiation temperature T = 1350 °C) in comparison to modified STRESS code prediction with corrosion law equ.(2)