THE INFLUENCE OF THE STATISTICAL SCATTER OF BOTH THE GEOMETRY PARAMETERS AND THE COATING STRENGTH ON THE FAILURE RATE OF COATED FUEL PARTICLES

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SUMMARY

An HTR fuel particle consists of a spherical pyrocarbon (PyC) and siliconcarbide (SiC) coated fuel kernel. The kernel diameter and the thickness of the adjacent buffer coating layer, a very porous PyC which provides free volume for the fission gases, have a significant influence on the stresses generated by the fission gas in the outer coating layers, which may consist of two dense PyC layers with a SiC interlayer. These outer layers form the pressure vessel for the retention of fission gases and provide a diffusion barrier for the solid fission products. The diameter of the kernel is 200–800 μm and the entire coating thickness is about 150 μm. The particle must be designed such that the stresses in the coatings, which increase with irridation time, do not exceed the coating strength in order to avoid breakage of the coating and release of fission products.

The study was performed for particles with a SiC interlayer. Here the whole fission gas pressure acts on the SiC as its Young's modulus is about ten times higher than that of the two dense PyC layers. This stress is reduced by the irradiation induced shrinkage and creep of the two dense PyC layers. It can be determined with stress model calculations taking into account the mechanisms mentioned above. It is assumed that the particle fails when the SiC breaks. Calculations show that the influence of the outer coating layer thicknesses on the SiC stress is far below that of the kernel diameter and buffer thickness.

The kernel diameter and buffer thickness have a scatter which is determined by the fabrication process, for this scatter a Gaussian distribution was assumed, the mean values and standard deviations were measured. The SiC strength shows an additional material inherent scatter for which a Weibull distribution was assumed. Its mean value and Weibull parameter were measured. It can be shown that the influence of the scatter in the geometry parameters on the mean value of the SiC strength may be neglected.

Stress model calculations were performed systematically for various buffer thickness and kernel diameter combinations at different burn-ups. Using the calculated stresses and the distributions of these two parameters it is possible to calculate the stress distributions over the particles at the corresponding burn-ups. Taking into account the strength distribution of the SiC, the fraction of particles expected to break was then determined as a function of burn-up.

1. Introduction

The fuel particles of the HTR consist of small spherical pyrocarbon (PyC) and siliconcarbide (SiC) coated fuel kernels. High stresses may be induced into the coatings during irradiation due to the fission gas pressure, the irradiation induced dimensional changes of the PyC layers and the kernel swelling.

In order to optimize the fuel particle with respect to its mechanical performance it is desirable to have a model which calculates the stresses in the coatings during irradiation. Several models [1] [2] have been established for this purpose. The model which is used at KFA was established by H. Walther [3] [4]. It calculates the stresses in the different coating layers of a particle with PyC coating only (biso) as well as a particle with an additional SiC interlayer (triso).

The stress in a certain particle type can be calculated at each interval during irradiation, but it must not represent the stresses in all particles of that type. This is so because there may be a considerable scatter in some geometry parameters, for instance, which leads to a stress distribution in the particles. Therefore, a fraction of the particles may have stresses much higher than the reference particle (particle with mean values for all parameters), these particles may fail earlier in irradiation than the reference particle.

During irradiation of coated particles, their fission gas release normally remains at a level of R/B[+)] = 10^{-5}, mainly caused by the contamination with fissile material. It may increase at high burn-ups. If this increase is caused by breakage of the coatings, when the fission gas pressure becomes too high, then the start of the increase and its rate can be determined by a calculation of the failure rate of the particles using the stress model.

The model can be used, for instance, to calculate the stresses in the coatings of a particle at a certain burn-up as a function of its geometry parameters. As these are more or less statistically distributed around their mean values, a statistical distribution of the stresses around those of the reference particle will result. If this distribution has a scatter that is high enough, a fraction of the particles may start to fail earlier than the reference particle.

The calculated statistical distributions of stresses resulting from the distribution of one parameter have a high scatter if both of the following criteria are fulfilled:

a) The calculated stress depends significantly on this parameter.

b) The value of the measured ratio between standard deviation and mean value of this parameter is high.

If there are more parameters for which (a) and (b) are fulfilled, the resulting scatter in the stress distribution is increased. The stress distribution can be calculated under the assumption of a Gaussian distribution for each of the parameters regarded.

The statistical distribution of stresses over the particles has been calculated for triso

[+)] R/B is the fraction : fission gas released/fission gas produced

particles for various burn-ups, taking into account two parameters: kernel diameter and buffer layer thickness. Other parameters such as the SiC layer thickness and the thickness of the inner and outer dense PyC layer, need not be regarded here, since their influence on the SiC stress and their statistical scatter are relatively low.
With this statistical distribution of stresses, the failure rate was determined:
1) assuming that the strength of the SiC has a difined value /8/ /12/
2) under the improved assumption that the strength of the SiC, being a brittle material, has a statistical distribution according to Weibull /11/.

2. The stress model used

A discription of the stress model was given at the first SMiRT conference /6/. The model has been successfully applied to both, biso and triso particles.
The validity of this model for triso and biso particles has been tested by means of an irradiation experiment especially designed for this purpose /5/ /7/.

3. The method of predicting the failure rate; influence of the statistical distributions of significant fabrication parameters and of the material strengths

In this paper, we partly follow a study referred to under /8/. In the case of triso particles, there are two geometry parameters which significantly influence the stress generated in the SiC layer. These are: the buffer thickness and the kernel diameter /9/. As the SiC has a Young's modulus and strength much higher than PyC /10/ /11/, the fission gas pressure and kernel swelling induce stresses mainly into the SiC layer, and it is therefore the SiC which, in case of a rupture, breaks first; the PyC layers then break immediately afterwards because of their lower strength. In this connection, we must presume that the two PyC layers do not break earlier than the SiC because of their own irradiation induced shrinkage.+) Thus, in the case of a triso particle, it is the stress in the SiC which governs the particle behaviour. The kernel diameter and buffer thickness are statistically distributed. In the analysis, a Gaussian distribution H_x is assumed for the kernel diameter x. For the buffer thickness y, the distribution is H_y. The mean values and standard deviations of x and y are determined for a certain particle type by measuring x and y on an adequate number of particles.

Now a parametric study of the stresses in the SiC layer is performed using the stress model. For the calculation, x and y are varied symmetrically around their mean values. The resulting stress in the SiC at the end or any point during irradiation can be marked in an x-y plot. For Stresses of 0, 100, 200 MN/m^2, etc., iso-stress lines can be found by interpolation in this plot. We call these lines $y = Y_{\sigma_i}(x)$. All particles located on or below the lines $Y_{\sigma_i}(x)$ have buffer thickness equal to or below $Y_{\sigma_i}(x)$ and, thus stresses in the SiC $\geq \sigma_i$. The cumulative probability W_i (σ_i) of having particles whith stresses $\geq \sigma_i$ in the SiC is therfore given by

$$W_i(\sigma_i) = \int_0^\infty H_x \left(\int_0^{Y_{\sigma_i}(x)} H_y \, dy \right) dx \qquad (1)$$

+) This may happen as is shown in Ref. /11/.

The calculated points W_i (σ_i) can be plotted in a logarithmic scale as is shown in Fig. 1. A polynomial of second order can be fitted well to these ponts, giving the function w (σ). This curve is also shown in Fig. 1.

3.1 Assumption that the strength of the SiC has one definite value

When calculating the fraction of particles that failed it has been assumed up to now that the SiC has a well defined strength /8/ /12/, and that the fraction of particles having stresses $\geq$200 MN/m^2 should break.

3.2 Assumption that the strength of the SiC is statistically distributed according to Weibull

Measurements performed on SiC /11/ /13/ indicate that its strength has a material inherent scatter. It was assumed that this scatter can be described by Weibull statistics, formulated as follows:

$$P(\sigma) = 1 - \exp\left[\ln 0.5\left(\frac{\sigma}{\bar{\bar{\sigma}}}\right)^m\right] \qquad (2)$$

P (σ) is the cumulative probability of having a strength $\geq\sigma$. m is the 'Weibull parameter', depending on the width of the distribution. $\bar{\bar{\sigma}}$ is the median strength which, in practical application, can be put equal to the mean strength /13/. The probability density of the strength distribution H(σ) is the derivative of equ. (2). If we multiply the cumulative probability w(σ) of having stresses $\geq\sigma$, which is obtained by the fit in Fig. 1, with the probability density H (σ) for strengths = σ and integrate over σ , we get an improved expression for the fraction of broken particles F:

$$F = \int_0^\infty H(\sigma)\, w(\sigma)\, d\sigma \qquad (3)$$

3.3 Determination of the dependence of failure rate on burn-up

For triso particles, the functions $Y_{\sigma_i}(x)$ are determined for different burn-ups and the values of W_i (σ_i) are calculated with equ. (1):

From the fitted curves w (σ) shown in Fig. 1, the number of particles having stresses above a defined value for the SiC, say 200 MN/m^2, can be determined for each burn-up. On the other hand, if a statistical distribution according to weibull H (σ) is assumed for the strength of the coating shell the fraction of broken particles can be determined with equ. (3).

4. Calculations performed with the stress model

An irradiation test BR2-P21 is planned for triso feed particles (EC 658/659) and triso breed particles (EO 819/820). For these particles, stress model parametric studies and calculations as described in Chapter 3 were carried out.

4.1 The feed particle EC 658/659

The measured mean values for the geometry of this particle are given in Table I.

Fig. 2 shows the stress distributions in the SiC layer for the end burn-up. The ratio between standard deviation and mean value of the buffer layer thickness is varied in this plot from 10 % (~value measured on the particle) to 15 % and 20 %. As can be seen, the thickness distribution of the buffer layer has a significant influence on the stress distribution. The influence increases rapidly with increasing stress. Fig. 3 shows the stress distribution in the particles for the end burn-up. The ratio between standard deviation and mean value of the kernel diameter is varied in this plot. It has the values 5 % (~value which was measured on the particles) and 10 %. When comparing Fig. 2 with Fig. 3 it becomes evident that the influence of the scatter in the buffer layer thickness on the stress distribution in this particle type is much higher than that of the scatter in the kernel diameter distribution.

In Fig. 4 the fraction of particles having stresses in the SiC layer above 200 MN/m^2 is plotted versus burn-up (curve a) as well as the fraction of broken particles assuming a Weibull distribution for the strength of the SiC with $\bar{\sigma}$ = 350 MN/m^2 and m = 7 (curve b). During the whole irradiation process, curve b remains by a factor of ~10 below curve a. At end burn-up, they reach values of R/B = 10^{-5} and 10^{-6}, respectively. Curve c on this plot shows the fraction of broken particles on the assumption that the strength of the SiC has the defined value σ= 350 MN/m^2 (mean strength of SiC measured) and all particles having stresses in the SiC layer $\geqslant\sigma$ would break. This curve remains by more than one order of magnitude below curve b. A comparison between curve b and curve c shows that the scatter in the SiC strength can have a significant influence on the number of broken particles.

4.2 The breed particle EO 819/820

The measured mean values for the geometry of this particle are given in Table II.

In Fig. 5, curve a shows the fraction of particles having stresses $\geqslant$200 MN/m^2, while curve b gives the fraction of broken particles on the assumption of a Weibull distribution for the strength of the SiC, plotted versus burn-up. The assumption of a defined strength of the SiC = 200 MN/m^2 leads to values which are by a factor of 5 above those where a Weibull distribution is assumed for the strength with $\bar{\sigma}$ = 350 MN/m^2 and m = 7. Curve c shows the fraction of particles that break if the SiC has the defined strength of σ = 350 MN/m^2 (mean value for SiC strength measured). Curve b and c are relatively close to each other, which leads to the conclusion that here the scatter in the SiC strength is not so significant for the breakage rate as in the case of particle type EC 658/659, as was shown in 4.1.

For the end burn-up, the fraction of broken particles according to curve a is 10^{-2}, while, according to curve b, it is lower by a factor of ~10.

Conclusions

As should be shown, the statistical scatter of the buffer layer and the kernel diameter play an important role for the stress distribution in the SiC shell over the triso particles during irradiation. And it is the stress in the SiC of this particle type which governs its irradiation behaviour. The scatters of the other parameters (inner PyC layer thickness, and outer PyC layer thickness) as well as their influence on the SiC stress /9/ are small enough to be neglected here. The spread in the buffer layer thickness is the most important factor for the stress distributions. The fraction of broken feed particles remains always below that of the breed particles. However, the stresses in the SiC in the reference feed particle at end burn-up are higher than those in the reference breed particle. The reason for this is that the value of the ratio between standard deviation of the buffer and the mean value is by a factor of 2 higher for the breed particles compared to that for the feed particles. On the assumption of a Weibull distribution for the SiC strength, the number of broken feed particles should be $\sim 10^{-6}$ at the end of irradiation. The fraction of broken triso breed particles at end burn-up should be 10^{-2} if failure is assumed to take place when the stress in the SiC is higher than 200 MN/m^2. The fraction of broken feed particles would then be $\sim 10^{-5}$. If a realistic Weibull distribution of the SiC strength (mean SiC strength $\bar{\sigma}$ = 350 MN/m^2, Weibull parameter m = 7) is assumed, the fraction of breaking particles should be lower by a factor of 10 for both particle types examined. It could be shown that the scatter of the SiC strength, taken as a Weibull distribution, leads to a significantly higher fraction of broken particles than the assumption of a defined SiC strength of σ = 350 MN/m^2 in the case of particle type EC 658/659. This effect was much less pronounced in the case of particle type EO 819/820.

These predictions of the model will be checked by the irradiation experiment BR2-P21.

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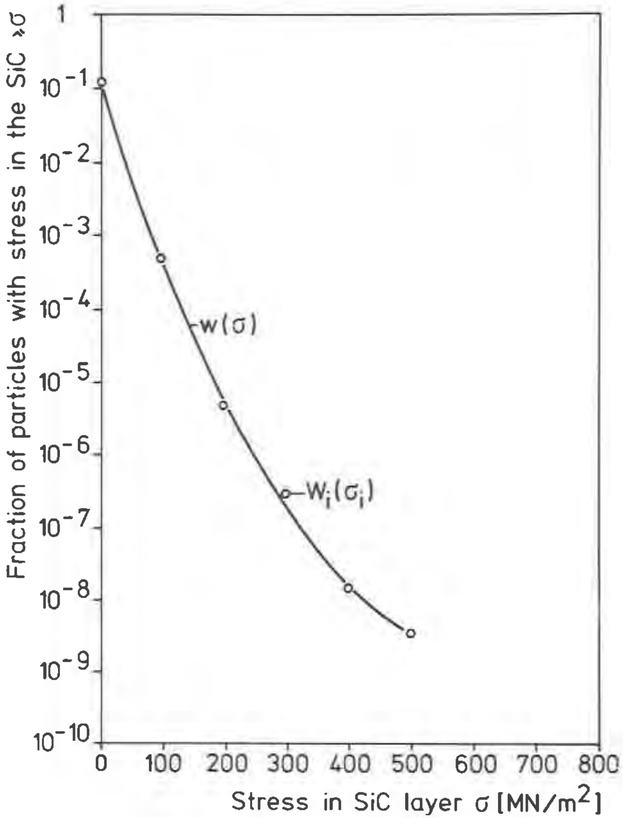
Table I: Measured mean values for the geometry of particle EC 658/659

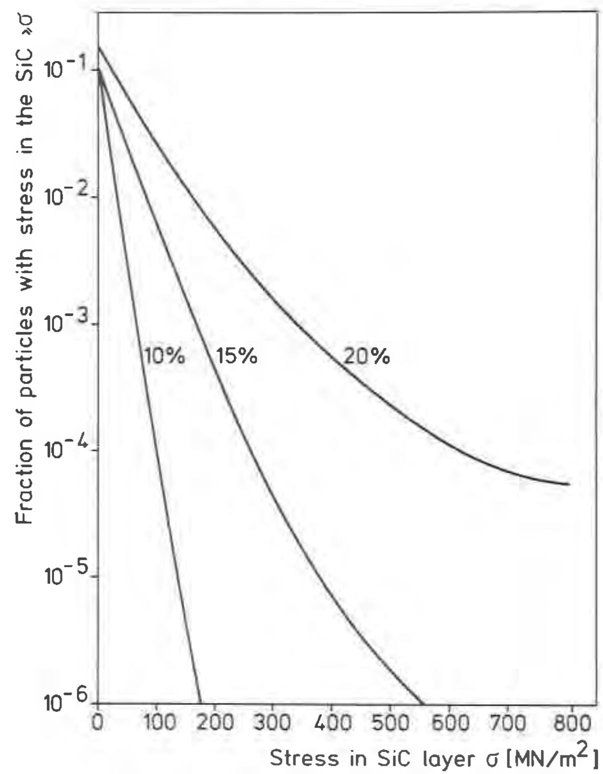
Kernel	
Diameter (μm)	196
Standard Deviation (μm)	11
Buffer Layer	
Thickness (μm)	95
Standard Deviation (μm)	11
Inner PyC Layer	
Thickness (μm)	31
Standard Deviation (μm)	2.9
SiC Layer	
Thickness (μm)	25
Standard Deviation (μm)	1.9
Outer PyC Layer	
Thickness (μm)	38
Standard Deviation (μm)	5.5

Table II: Measured mean values for the geometry of particle type EO 819/820

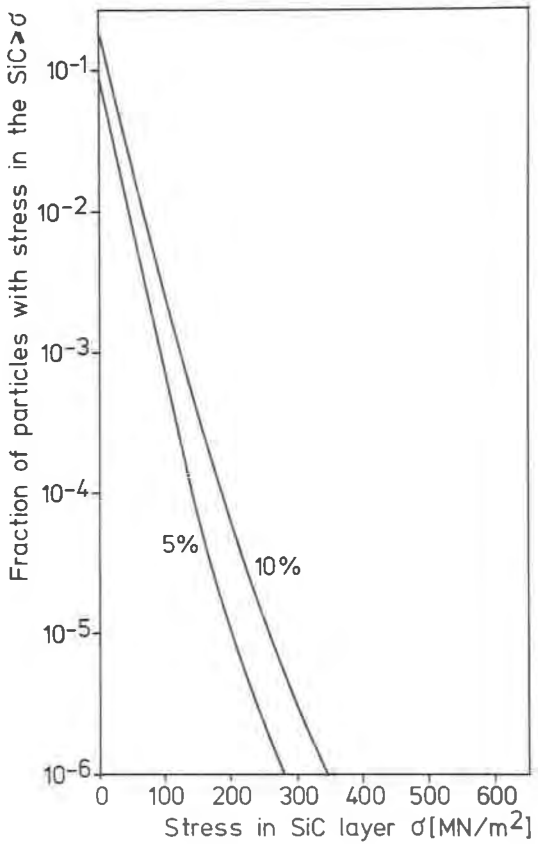
Kernel	
Diameter (μm)	496
Standard Deviation (μm)	6.9
Buffer Layer	
Thickness (μm)	66
Standard Deviation (μm)	12.9
Inner PyC Layer	
Thickness (μm)	37
Standard Deviation (μm)	4.6
SiC Layer	
Thickness (μm)	33
Standard Deviation (μm)	1.9
Outer PyC Layer	
Thickness (μm)	34
Standard Deviation (μm)	3.9



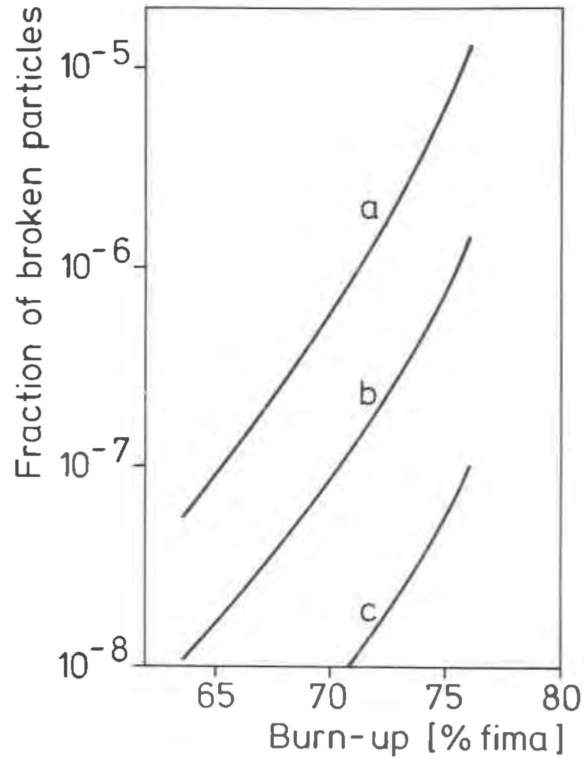
1: Calculated cumulative probabilities $W_i(\sigma_i)$ for stresses in SiC-layer $\geq\sigma_i$ of particle type EC 658/659. A second order polynomial $w(\sigma)$ fitting the data is shown.



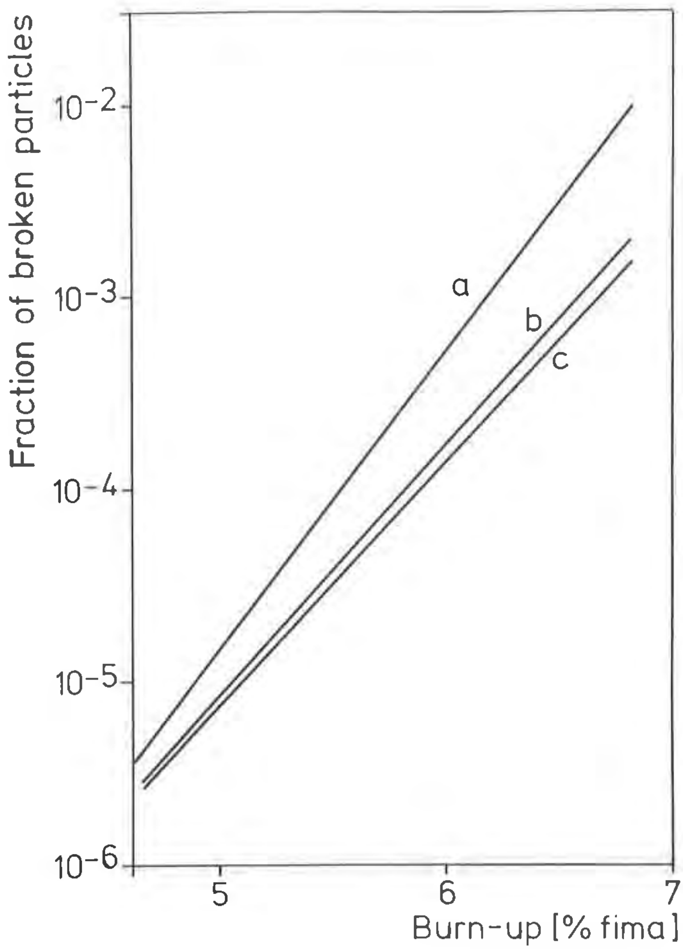
2: Fraction of particles with stress in the SiC layer $\geq\sigma$ plotted versus σ for the end burn-up of 76 % fima. The ratio between standard deviation and mean value of the buffer layer thickness is varied: 10, 15 and 20 %. Particle type EC 658/659.



3: Fraction of particles with stress in the SiC layer $\geq \sigma$ plotted versus σ for the end burn-up. The ratio between standard deviation and mean value of the kernel diameter is varied: 5 % and 10 %. Particle type EC 658/659.



4: Fraction of particles with stress in the SiC layer ≥ 200 MN/m^2 (curve a), fraction of broken particles assuming a Weibull distribution for the SiC strength, with $\bar{\sigma}$ = 350 MN/m^2 and m = 7 (curve b) and fraction of particles having stress ≥ 350 MN/m^2 in the SiC layer (curve c), plotted versus burn-up. Particle type EC 658/659.



5: Fraction of particles with stress in the SiC layer $\geqslant$ 200 MN/m^2 (curve a), fraction of broken particles assuming a Weibull distribution for the SiC strength, with $\tilde{\sigma}$ = 350 MN/m^2 and m = 7 (curve b), and fraction of particles having stress $\geqslant$350 MN/m^2 in the SiC layer (curve c), plotted versus burn-up. Particle type EC 819/820.