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EXPERIMENTAL VERIFICATION OF STRESS MODEL CALCULATIONS FOR HTR FUEL PARTICLES

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Summary

The fissile and fertile material for gas cooled high temperature reactors is contained in microspheres coated with several pyrocarbon (PyC) and silicon carbide (SiC) shells. The basic performance-limiting phenomenon in fuel particles is pressure vessel failure. The effects of irradiation temperature, neutron exposure, kernel burn-up and kernel and coating dimensions and densities on PyC and SiC coating layer stresses can be estimated using coated particle stress analysis codes. The reliability of these calculations have to be proven by experimental verification. A new method was established for calculating failure rates directly for an unlimited number of particle parameter having a statistical distribution using a simplified time saving version of the Walther model. Material properties and geometry data from kernel and coatings are used as input parameters. For the kernel, Young's modulus, and fuel swelling are constant whereas fission gas release from the kernel is a function of burnup. For the coatings Young's modulus, Poisson ratio, irradiation induced creep coefficient, Poisson ratio in creep, irradiation induced unrestrained dimensional change rate in tangential directions are treated as function of neutron fluence. Material strength in tangential direction is treated as constant. From the calculated stresses and reasonable failure criteria for PyC and SiC the number of defective particles can be predicted for irradiation experiments. Experimentally the number of failed particles can be determined by two different methods. On the one hand the end of life fission gas release is measured and from this the number of failed particles can be estimated. On the other hand the whole fuel bodies are deconsolidated and the number of failed particles is determined. To reduce the number of free parameters which can be used to fit the stress code, properties like fuel swelling, change in optical anisotropy, end of life internal gas pressure, SiC-strength, distribution of bufferlayer thickness and kernel diameter were determined experimentally and the corresponding input data computed. The calculations agreed sufficiently with the experimental results from irradiation tests.

1. Introduction

In all prototypeand large high temperature gas cooled reactors (HTGR) currently in service, under construction or planned the fuel is inserted as coated fuel particles embedded in a graphitic matrix which forms the fuel elements. This is to include a very small amount of fuel into a so called pressure vessel providing an excellent retention for gaseous and solid fission products.

For accident analysis, siting dose calculations and design values for sizing plant components several fission product release models for large HTGR have been proposed. A fission product release model include usually

- fuel particle failure
- release from fuel particles
- transport through matrix
- release to the coolant gas

Fuel particle failure is one of the most complex portion of the overall release model and only this part will be considered in the following presentation.

Four main phenomena contributing to coating failure have been identified so far

- pressure vessel failure due to radiation induced stresses and internal fission gas pressure
- unidirectional migration of kernel material into the coating
- fission product attack of the siliconcarbide layer
- imperfect as -fabricated coatings.

Besides fabrication defects there is essentially just one dominant basic fuel failure and this is the pressure vessel failure. It occurs when the stresses induced in the coatings exceed their strength.

To predict the number of coated fuel particles failed by pressure vessel damage several analytical models have been developed and improved by Walther[1], Bongartz [2], Martin [3] and Kaae [4]. All this models can be used in principle for particles with a pure pyrocarbon coating as well as with pyrocarbon (PyC) coatings improved by an additional silicon carbide layer.

The objectives of this presentation are to show the suitability of a selected model for designing particles and the experimental verification of the model with regard to several assumptions which where made to fit the model to experimental results.

2. HTR-fuel particle design

The general design of a coated fuel particle is given by a kernel containing the fissile or fertile material covered by a porous buffer layer, which accomodates for kernel swelling, fission products recoil and for providing void volume for the gaseous fission products. This layer is covered by one or more dense layers consisting of PyC only (BISO-coating) or a SiC layer sandwiched between two PyC layers (TRISO-coating) which form the pressure vessel for the gaseous as well as a diffusion barrier for the solid fission products.

For the present HTR's besides a 400 μm $(Th,U)O_2$-BISO particle for the mixed oxide system three TRISO coated particles were developed: a 500 μm $(Th,U)O_2$-TRISO particle also for the mixed oxide system and a 200 μm UCO-TRISO particle together with a 500 μm ThO_2 TRISO particle for the feed breed system.
In all cases particle coating designs have evolved as a result of manufacture experience and irradiation testing coupled with the development and application of analytical stress models.

3. The analytical model

Historically the stress model used at KFA is based on the Walther-Stress model. Together with a statistical model based on the ideas of Gulden [5] it was applicable in a twofold manner. On the one hand it is used for designing particles by comparing apparent stresses which differ due the change of input parameters. On the other hand it is able to predict numbers of failed particles by using an appropriate failure criterion and the best available materials data. As it can be understood easily the quality of both results is different, the qualitative comparison of particles with different designs gives quite satisfactorily results whereas the quantitative calculations need to be improved regarding several input data.
During the last years it appeared that reliable values for particles can be obtained only by taking into account the scatter in the geometric data and other physical properties.
To account for this codes using the Monte Carlo Method were developed. However sensitivity studies and parameter variations as a base for designing coated particles were still a very time consuming procedure. Recently a new computer program was established by Bongartz [6] for calculating numbers of failed particles directly for an unlimited number of particle parameters having a statistical distribution. This can be done using a simplified version of the versatile but time consuming Walther model together with a Law of Error Propagation method. Since this code requires extremely low computation times it is a useful tool for iterative calculations for particle design optimization as well as for predicting even small numbers of failed particles.
Although this model is mathematically sophisticated the use as a quantitative model is still limited because of uncertainities in the input data e.g. creep constants or failure criteria for PyC, the mathematical formulation of the precise physical mechanisms is not completely solved e.g. influence of the increasing anisotropy on the shrinkage of pyrocarbon. To compensate for these uncertainties during calculating absolute numbers of failed particles the model had iteratively to be adjusted to results of irradiation tests on fuel particles.

4. Experimental verification of the model

The experimental verification of a model should include the adequate determination of the input data, sensitivity calculations for special parameters, identification of rupture criteria and finally, the comparison of predicted and measured results.

4.1 Determination of input data

Input data for the model used at KFA usually are divided in those which are assumed to change during irradiation with burn up, fast neutron fluence or temperature and those which are considered as constant. The latter can normally be determined by quality control or sophisticated laboratory experiments. The former can either be measured directly on irradiated particles or and this the most complicated case only be deduced with a physical hypothesis from measured data. A list of the main input data is given in table I.
Quality control of coated fuel particles is highly advanced because of the stringent requirements from reactor constructors, utilities and licensing authorities. All geometric data together with their standard deviations can be determined with sufficient accuracy by means of fully automated particle size analysers and X-ray microradiography. Recently a sophisticated method to determine the true densities and densitygradients of all pyrocarbon coatings on the as-fabricated coated particles was developed using the change in intensity of a soft X-ray passing through the coating layers. Anisotropy of the optical reflection which can be transferred into BAF (Bacon anisotropy factor) are also determined by a fully automated device. Other physical data like fracture strength of SiC and PyC are determined by sophisticated methods described by Bongartz [7].

An important in put parameter for the program is the neutron induced unrestrained dimensional change of the layers. Specially designed irradiation experiments yielded a lot of results but their remain still some doubts whether the data obtained from flat material can be used for spherical coatings. But so far no experiment was designed where spherical coating layers could be irradiated unrestrained. From our last experiments we assume that the buffer layer shrinks very fast and isotropically where the shrinkage of the high dense layers depends on their density.
From our last high burn-up experiments also the dimensional behaviour of different fuel kernel material could be deduced. It was found that swelling of UO_2 kernels is about twice as high as swelling of UC_2 or UCO-kernels. At very high burn-up saturation takes place. Neutron induced changes in anisotropy were also observed by measuring anisotropy on irradiated particles. They indicated that layers deposited from propene and ethine (LTI) show much higher stability against fast neutron flux than coatings from methane (HTI) do.
The amount of fission gases and CO which are liberated during irradiation from the kernel can be measured by hot crushing of individual particles and determining the amount of Xe, Kr and CO simultaneously with a quadrupol mass-spectrometer. These results revealed that in particles with oxidic kernels after high burn up the CO pressure can be about 50 % of the overall gas pressure. This can be avoided by using pure UC_2 or only some 10 % of UC together with UO_2.

4.2 Sensitivity calculations

The analytical models indicate that fuel kernel diameter and buffer coating thickness control the fission gas pressure within a TRISO-coated particle. For TRISO-particles the internal pressure is the dominant factor in controlling the performance. We can show that with our model a decrease in the standard deviation of the buffer layer thickness of 30 % results in a decrease of the number of failed particles by more than two orders of magnitude. Similar results are obtained by changing kernel diameter standard deviation or inner high dense PyC layer thickness. All these predictions agree rather well with results from irradiation experiments although the quantitative numbers will be different. However the influence of irradiation temperature is not described properly with the present input data. From the model only ten or twenty percent increase in the number of failed particles are predicted for an increase in temperature from 1100 to 1300°C whereas recent irradiation test show clearly differences of one order of magnitude and more. We attribute this to the unknown temperature dependence of some main input parameters as dimensional changes and creep coefficient. Here is further work needed to improve the input parameters.

4.3 Identification of rupture criteria

For design calculations it is sufficient to compare the stresses in different layers depending on the desired parameter variation. To predict the number of failed particles however rupture criteria are necessary.
Different laboratories have experimentally determined the maximum strength of SiC-layers and its distribution. It is rather well agreed, that SiC has a median strength of 350 M Pa and a Weibull parameter of about 7. All our failed TRISO particles are calculated with these numbers.

Besides the not entirely understood failure mechanisms also the mean strength of PyC is not well established. This stems mainly from the large scatter in manufacture conditions and materials data. The rupture criterion can be defined as a mean strength together with a Weibull parameter or a limiting hoop strain. For our calculations we used for

Buffer : median strength 100 MPa
Dense PyC: median strength 200 MPa
Weibull parameter 4

Table 1: Input parameter for the analytical model

Pre - irradiation data		Data from irradiation experiments	
Input parameter	Method of determination	Input parameter	Method of determination
1. Geometrical data: . Kernel : Diameter standard dev. . Buffer : Thickness standard dev. . Dense PyC: Thickness standard dev. . SiC : Thickness standard dev.	Quality control - particle size analyser - microradiografy	1. Irradiation History . temperature . fast fluence . burn-up	Capsule instrumentation - thermocouples - monitor wires - SPN-detectors
2. Physical data: . Density . Anisotropy of PyC . Mean strength of PyC and SiC in tangential direction . Weibull parameter . Young's modulus . Poisson's ratio	Quality control - X-ray intensity - Hg-Pyknometry - gradient column - optical anisotropy Laboratory experiments - brittle ring test - deformation of particles	2. Physical data . kernel swelling . Neutron induced dimensional change of porous and high dense pyrocarbon . creep coefficient Poissons ration in creep . Neutron induced changes in anisotropy	Post irradiation examination - X-ray microradiografy - dimensional measurements - optical anisotropy
3. Chemical data: . Kernel composition	Quality control - wet chemistry	3. Chemical data . fission gas release from kernel . oxygen per fission	Post irradiation examination - cracking of irradiated kernels and determination of fission products - Determination of CO in the void volume

4.4 Comparison of predicted and experimentally determined results

4.4.1 The irradiation experiment

Coated fuel particles from one standard quality are embedded in the original matrix of the fuel elements in form of coupons, compacts or spheres. These samples are then inserted in a fully instrumented and swept capsule which is subsequently irradiated in a rig-type device in a material test reactor.
During the irradiation temperature, thermal and fast flux and fission gas release is monitored continuously.
After irradiation the rig is dismantled, the samples are deconsolidated and the number of defective particles is determined on a representative aliquot.

4.4.2 Experimental determination of defective particles

During irradiation the fission gas release shows clearly the behaviour of the coated fuel particles. The basic release comes from the contamination which is present at a low level. R/B-values less than 10^{-5} usually are produced by contamination. Sharp rises during irradiation or continuos growing of the fission gas release signalize the onset of particle failure. From out of pile measurements for all R/B values following a proposal of Myers [8] failed particles can be determined with regard to the increasing kernel release with growing burnup. So the fission gas release gives not only the final number of failed particles but also the time dependence of the failure mechanism.

After irradiation failed particles can be determined roughly with ceramographic sectioning. This is sufficiently accurate if more than 10 % failed. For failure fraction below 1 % more sophisticated methods have to be used to investigate large numbers of loose particles. Today two methods can be applied to KFA particles. The one is the IMGA (irradiated microsphere gammaradiation analyzer)-system the other is the PIAA (post irradiation annealing and β-autoradiography)-system.

4.4.3 Comparison of calculated and experimentally determined defective particles

To show the whole range in which the model is applicable we have selected two irradiation experiments. One experiment contained fissile particles which reached their final burnup of more than 70 % fima in about 510 equiv. full power days (fig. 1). Only at the very end of the experiment R/B-values increased over 10^{-5} which may be the first sign for particle failure. The model shows that theoretically the failure onset should be still more postponed. We assume that at these high burn ups also other failure mechanisms may contribute besides the pressure vessel failure. Irradiation temperature was kept constant at 1200^{0}C.
The other experiment contained 500 μm fertile particles with 4 % uranium to increase slightly the burn up. In this case irradiation took place in two different capsules at 1100^{0} and 1300^{0} C.
The irradiation time was about 380 equiv. full power days and the maximum burn up about 10 %

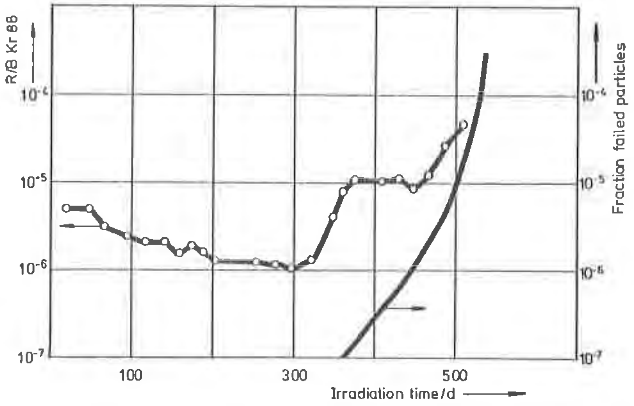

Fig. 1: Comparison of measured R/B values with predicted failure for 200 µm UC$_2$-fissile particles

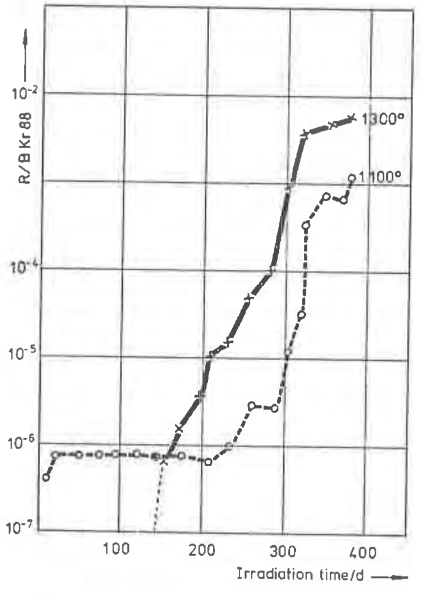

Fig. 2: Fission gas release from 500 µm fertile particles at different irradiation temperatures

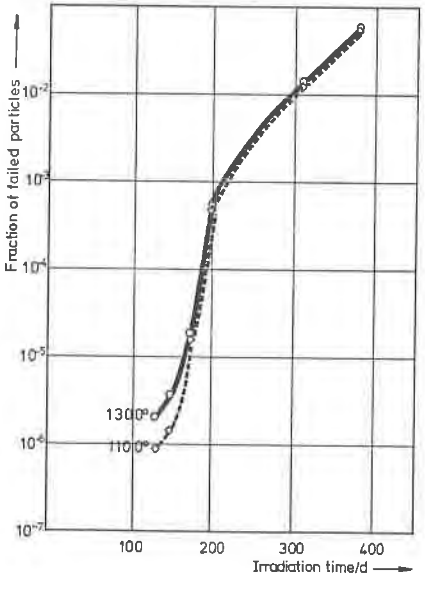

Fig. 3: Calculated numbers of failed particles for two different irradiation temperatures

fima (fig. 2). Today we know, that the buffer layer was under-designed and the number of failed particles exceeded the specified level. However this gave us the possibility to observe fission gas release over several orders of magnitude. The comparison between model prediction (fig. 3) and experiment (fig. 2) shows first that the temperature dependence of the failure mechanism is not well fitted in the model. On the other hand the onset of particle failure as well as the sharp rise in the fraction of failed particles is described very well. The difference in the values of failed particles and the R/B-values comes from the fact, that fertile kernels with 10 % fima only have a substantial kernel retention for fission gases. This is in agreement with observations from other experiments.

5. Conclusions

The stress model used at present at KFA together with a statistical program based on the Law of Error propagation can be applied for designing all present reference particles because of very short computation times. Together with experimentally determined input parameters also predictions for fraction of failed particles can be made in the whole range of diameters between fissile and fertile particles.
However further development is needed to describe more accurate the temperature dependence of failed particles. But with the data available a better fit seems possible.

6. References

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