BEHAVIOUR OF HTGR COATED PARTICLES AND FUEL ELEMENTS UNDER NORMAL AND ACCIDENT CONDITIONS

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Abstract

Main results of testing HTGR coated particles and spheric fuel elements developed in Scientific and Industrial Association "Lutch" under conditions of higher level of energy release and temperature than those designed are given in the report. The summarized data on tightness and characteristic defects change, on gas and solid fission products release under model accident conditions before, during and after radiation are presented.

1. INTRODUCTION

While studying possible HTGR accidents development three main reasons of their appearance can be distinguished:

a) Sharp introduction of a considerable positive reactivity to the reactor core increases the neutron flow density, energy release in fuel elements and then their temperature;

b) Consumption (decrease or discontinuation) of the coolant for heat removal from the core directly increases the fuel temperature;

c) Loss of tightness of the primary circuit results in ingress if a considerable quantity of air or water gets from the second circuit to the core with subsequent corrosion processes.

The present work is aimed at studying the first two items.

Table I

CHARACTERISTICS TESTING OF COATED PARTICLES AND FUEL ELEMENTS

Parameter	Size, μkm	Density, g/cm^3	Technological peculiarities
COATED PARTICLES			
Kernel UO_2	500±50	10,4-10,8	(a)slicker
Enrichment, of U-235- 21 and 45 %			(b)sol-gel
PyC	80-100	0,9-1,1	C_2H_2-Ar
PyC	15-20	1,5-1,6	In LTI is absent
PyC	30-50	1,8-1,9	(a) HTI (b) LTI
SiC	30-60	3,2	(a) $SiCl_4$ (b) CH_3SiCl_3
PyC	40-80	1,8-1,9	(a) HTI (b) LTI
FUEL ELEMENTS			
			CUP,graphite
Fuel zone	Ø50 mm	1,82-1,85	a) 30PG
Fuel Element	Ø60 mm	1,82-1,85	b) MPG-6
Charge U-235 in fuel elements - 0,5-1,5 g. Contamination U-235 - 10^{-7}- 10^{-9}g/g graphite.			

2. CHARACTERISTICS OF HTR FUEL

Coated particles (CP) and spheric fuel elements (FE) on their base, manufactured in Scientific Industrial Association (SIA) "Lutch" under laboratory conditions applicable to VGR-50, VG-400 and VGM designs were investigated. A detailed description of technological methods for manufacturing such FE was given in (1).

The peculiarity of the investigations was that they were performed parallely to the choice of construction, material content and development of technological conditions of FE production within the frame of the above mentioned designs. This is the reason of a wide range of thickness modifications of some CP coatings, pyrolysis gases and conditions of layer deposition from PyC and SiC, matrix graphite and others in different batches. They are given in the table and reflect the technology variants of 1981-1986. The main steps of the technological scheme were:

- manufacturing UO_2 kernels by the zol-gel and slicker spheroidization methods;
- deposition of protecting coatings in pebble-bed apparatus by pyrolysis of different hydrocarbons for creating pyrocarbon (PyC) layers and volatile silicon compounds to obtain a carbide layer (SiC);
- FE manufacturing by the method of carbonization under pressure from a mixture on the base of graphite powders and pitch binders with subsequent thermal and mechanical treatment;
- control operations;

In spite of a substantial number of construction and technology modifications, while CP and FE manufacturing, a big resemblance in their behaviour under test conditions was found in the research.

3. BEHAVIOUR OF UNIRRADIATED FUEL ELEMENTS DURING HIGH-TEMPERATURE TESTS

Material science aspects of CP behaviour in a free state after burnups at 1900-2000°C for 10-2000 hours and at 2250-2500°C up to 30 hours are discussed in (2) represented by S.Kurbakov at XI IAEA Conference on HTGR. CP characteristics are given in fig.1(b,c) and deal with activization of U and Si diffusion into pyrocarbon, SiC thermal decomposition with pore formation, mass losses and strength decrease at temperatures higher 2000°C. When the temperature increases up to 2500°C spalling of the outer PyC layer and through failures of the coatings are observed.

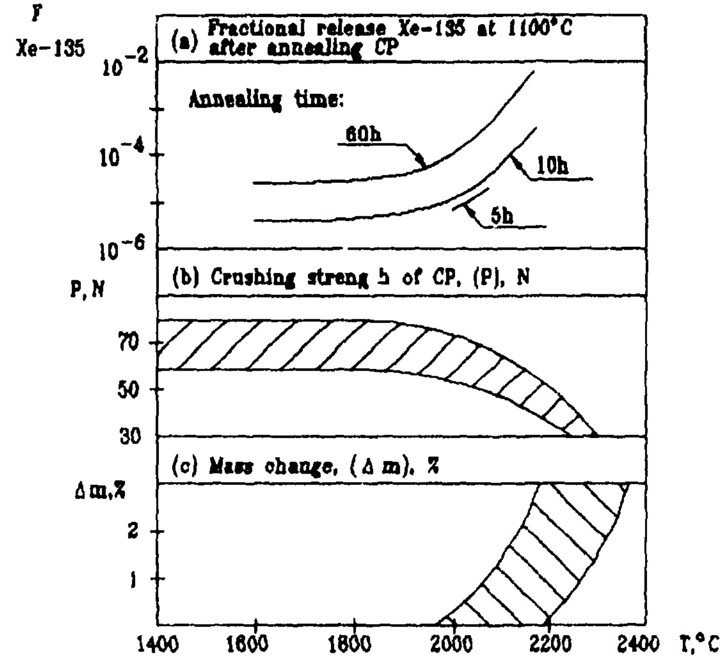

Fig.1. Change of unirradiated CP after annealing.

Besides that after a heating at different temperatures up to 60 hours CP tightness for gas fission products (GFP) by the "weak radiation" method in F-1 Reactor with subsequent annealing at 1100°C and Xe-135 release analysis was controlled (fig.1a)/3/. As we can see in the figure CP tightness also substantially decreases after the temperature of 2000°C is exceeded.

In FE the threshold temperature of coating failure beginning grows a little (2200-2300°C) due to banding effect of the matrix graphite. After FE annealing up to 2750°C for 1 min Xe-135 leakage at 1000-1100°C becomes equivalent to FE with kernels without coatings ($F \sim 2 \times 10^{-2}$) and after 15 min - to homogenious distribution of uranium ultra - dispersive powder in graphite ($F \sim 0,2$). A quick (15-30 s) FE heating from 2200-2500°C to 2700°C is accompanied by CP burst failure with FE fragments spalling.

4.GROWTH OF FE ENERGY RELEASE

An average energy release under design conditions is about 1 kW in one FE, the maximum value is up to 5 kW which corresponds to a specific power of 0.1-0.5 W in one CP. According to the calculation estimation in hypothetical accident situations, when positive reactivity is sharply introduced, energy release can increase by some orders in the fuel (4), that's why studying this factor influence becomes necessary. This factor is also important for interpretation of accelerated reactor tests results, where, as a rule, the specific energy release is higher than nominal.

Fig.2 illustrates the conditions of full-scale FE reactor tests with an increased energy release. FE behaviour peculiarities are:

Mark 2 - power is about 1 W per CP, irradiation time is 5-7 thousand effective hours. Critical fuel burnup (the beginning of the loss of tightness) is less than under nominal radiation conditions; the central pore is formed in some CP.

Mark 3 - power is up to 2 W per CP, irradiation time is up to 5 thousand effective hours. The "amoeba" effect becomes substantially active, the critical burnup is almost two times lower than the nominal one.

Mark 4 - power is up to 12 W per CP, irradiation time is 40 s. Irradiation rig was not equipped with a forced FE cooling system, that resulted in heating up to 2500-3000°C and in failure of both CP layers and all the FE. The FE imitator without CP retained its integrity. When the maximum temperature in such experiments doesn't exceed 2000°C the state of FE was quite satisfactory.

Mark 5 - impulse FE irradiation with fluence is $\sim 10^{17} cm^{-2}$, the maximum neutron flow density is $1.6\times10^{16} cm^{-2}s^{-1}$ and the impulse half-width is ~0.5s. FE were integral, GFP substantial release was not noticed, but detailed post-irradiation investigation was not performed. Such conditions of irradiation is maximum similar to HTGR - accident with a sharp introduction of positive reactivy.

Mark 6 - impulse (~1 μs) FE irradiation with fluence up to $10^{14} cm^{-2}$. GFP release increase was observed when the fluence was

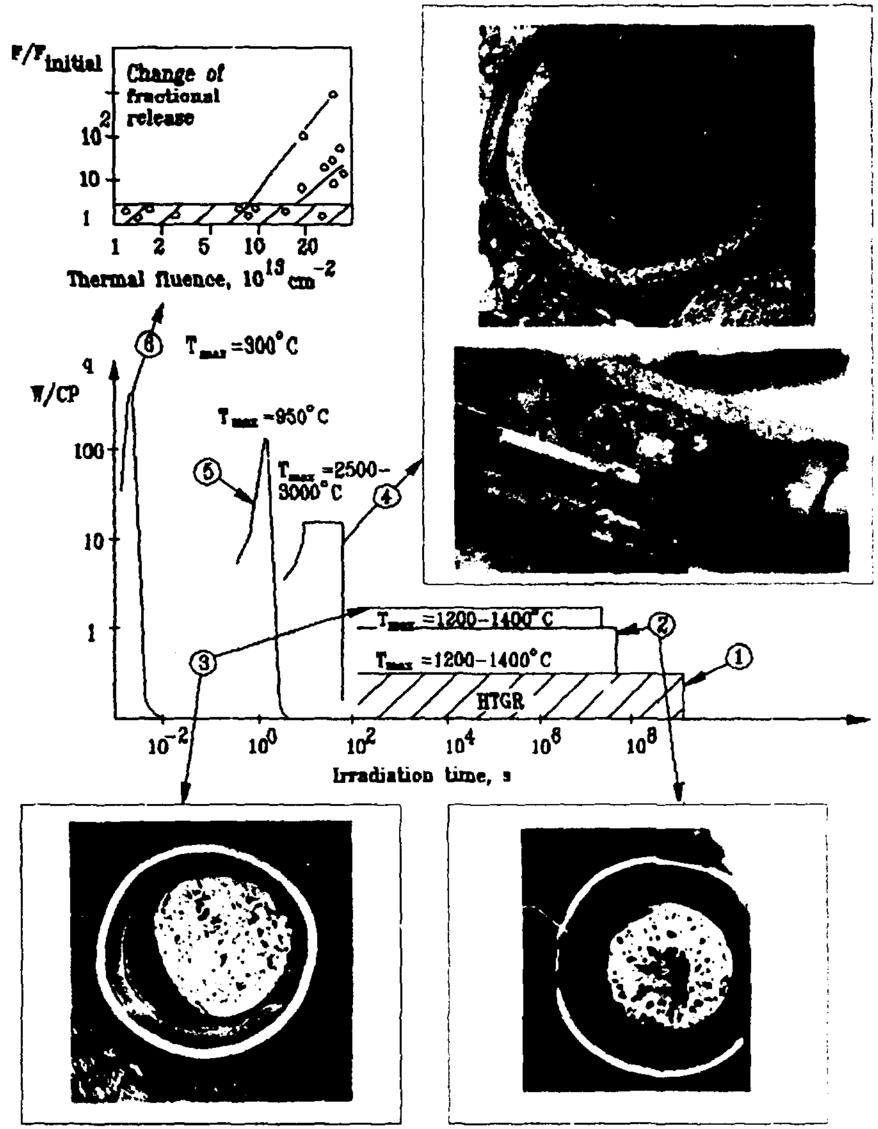

Fig.2. FE test conditions with a higher energy release in CP (q):
1 - HTGR; 2 - IVV-2M; 3 - SM-2; 4,5 - IGR; 6 - GIDRA

higher than $1\times10^{14}cm^{-2}$. It was found by the method of "weak irradiation" and post-annealing at 1100°C for Xe-135. SiC layer cracking was seen by the metallographic investigation. This result agrees well with Japanese specialists results and corresponds to the limit energy release about 1000 J/g of UO_2 (5).

5. HIGH-TEMPERATURE IRRADIATION

Investigations for founding out the limited possibilities of the protecting coatings were performed for CP in a free state or coupons content in IVV-2M Reactor.

The CP were irradiated in the temperature range 1000-2000°C up to a burnup about 5;10 and 15% *fima*, after that their tightness was controlled, at first the control of 20-40 irradiated CP was performed by the method of test annealing at 1000°C, and Kr-85 release was compared with kernels without coatings. Then the IMJA analysis of Cs/Ru release was done. In fig.3 the dotted line denotes CP service life zone, where numerous losses of tightness were not observed. Also one can see the structures characterizing CP state in different temperature-doze zones. It is remarkable that at 2000°C SiC porosity development (a), the process of kernel carbonization after coating destruction (b), SiC decomposition in zones with open cracks in the inner PyC layer at the increased irradiation temperature (c) take place. At lower temperatures the state of CP is quite satisfactory.

The zone of FE service life was investigated in the same reactor in the rigs of Vostok-type at the temperatures about 1000, 1200 and 1400°C (6). Sixteen FE of different types were irradiated in four rigs with autonomous selection of samples for studying ten isotopes of Xe and Kr content. This work allowed to establish the following (fig. 4):

- irradiation at the temperature 1000°C being normal for HTGR is accompanied with an insignificant growth of GFP losses, the critical burnup is higher than 15-20% *fima* (not obtained). R/B as a rule, is not higher than 10^{-6};

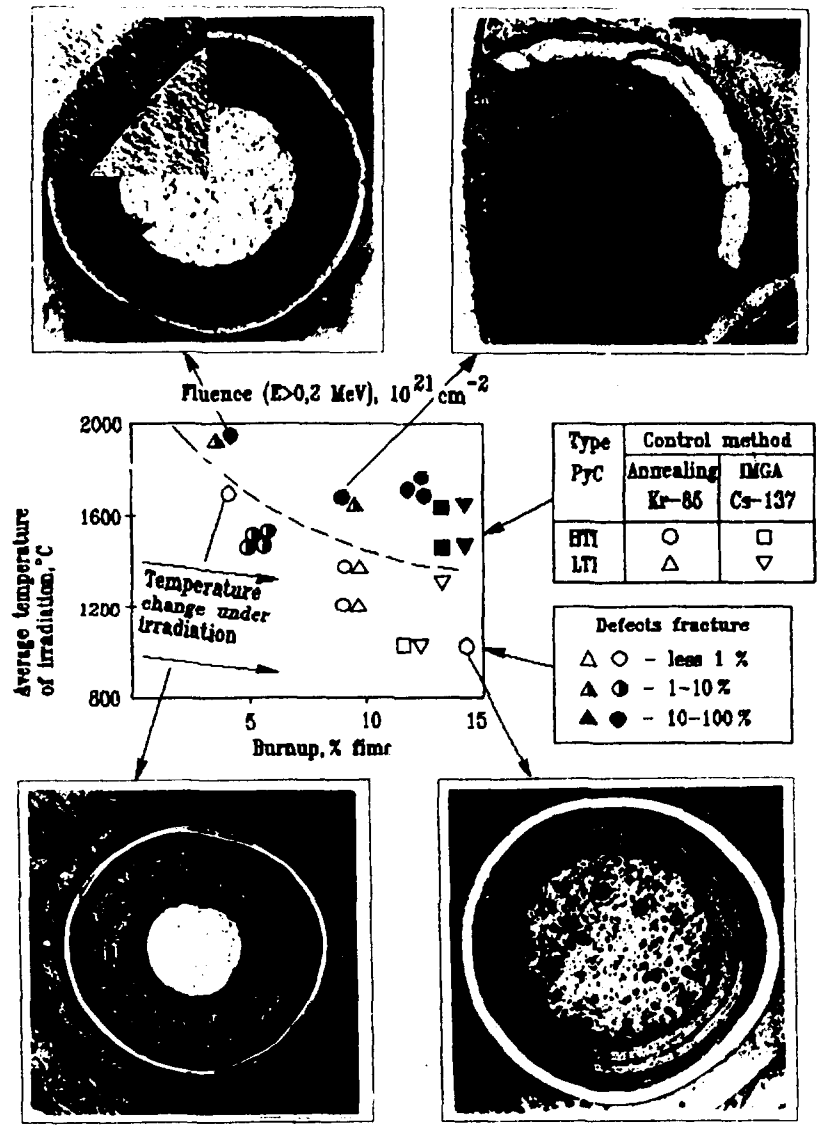

Fig.3. Tightness and a typical appearance of CP after irradiation at high temperatures in a coupon content.

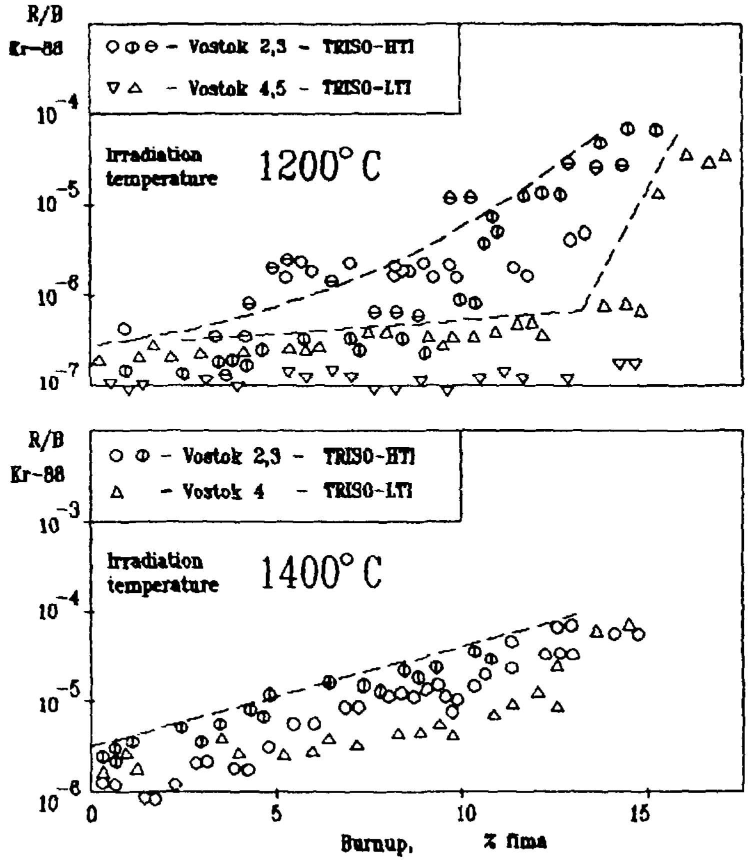

Fig.4. Fractional gas release from FE during prolonged irradiation at a higher temperature in IVV-2M Reactor.

- irradiation at 1200°C can be accompanied by loss of tightness of some CP after a burnup 10-15% *fima*, the limited values ($R/B=10^{-5}$) may be exceeded;

- irradiation at 1400°C is accompanied with GFP release growth up to $R/B = 10^{-5}$ at 5-13% *fima*.

The obtained results can characterize FE possibility to withstand a prolonged irradiation at higher temperatures, as a

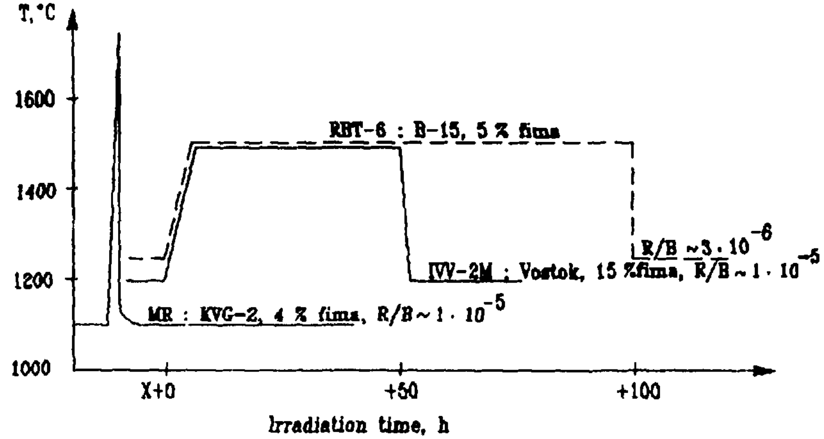

Fig.5. Test conditions of accident FE heating during service life irradiation.

result, for example, of active-zone thermal-physics calculation errors or FE technological defects. It should be taken into consideration that the tests in Vostok rigs are accelerated and service life estimations can be regarded as conservative.

In a number of service life reactor tests with FE after obtaining a burnup 5-10% *fima* a short-time temperature increase, imitating an accident cooling, was done (fig. 5). GFP release increase was not observed in any case after temperature return at the initial level.

6.POST-IRRADIATION INVESTIGATIONS

High-temperature tests of the irradiated CP are performed, as a rule, after FE desintegration. In this case defects appearance in CP coatings is possible at desintegration (especially chemical) and transportation, matrix banding also disappears.That's why Kr-85 release character can be various. It's shown in fig.6 for isochronic annealing with temperature growth

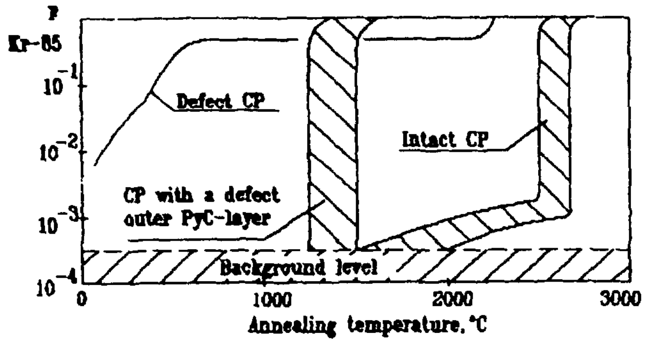

Fig.6. Typical fractional release (F) of irradiated CP.

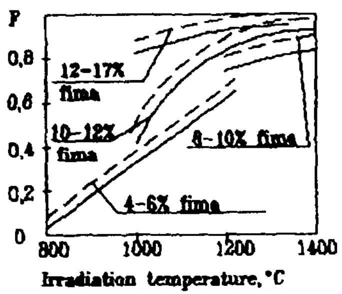

Fig.7. Fractional release of Cs-134(---) and Cs-137(-) from the kernel into the coating.

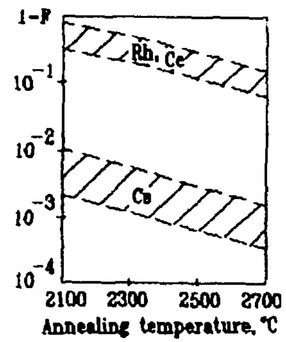

Fig.8. Residual content of solid fission products in CP after loss of tightness at annealing.

velocity about 2-6 K/s for intact CP, CP with defect outer PyC layer and defect CP.

While annealing intact CP up to 1900°C for 50h Kr-85 release was not observed in sensitivity limits ($\Delta F \sim 10^{-3}$). CP rejected after IMJA- analysis as unhermetical for caesium were "explosed" at 1300-1500°C with about 50% Kr-85 losses. This testifies to the fact that SiC properties concerning solid fission products retention can degradate when GFP tightness at a high fuel burnup (~20% *fima*) is retained.

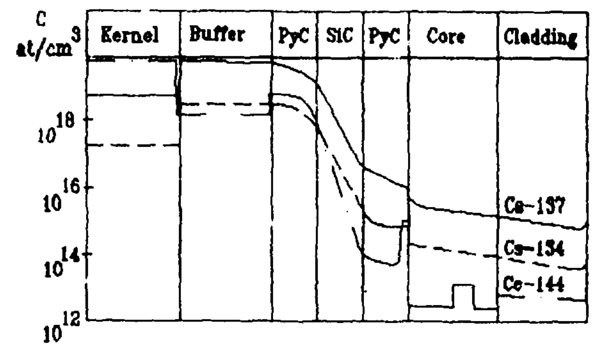

Fig.9. Concentration profiles (C) of solid fission products in FE after irradiation at 1200 C to 19% fima (IVV-2M, Vostok-4 fuel element 63).

The investigated results of caesium release from the irradiated kernel into the coating and residual content of Cs,Rh,Ce after annealing up to coating destruction are given in fig 7,8.

The matrix graphite is not a serious barrier for caesium release into the first circuit (fig.9) judging from the results of sampling by FE electrolitic desintegration and CP laser evaporation.

7.CONCLUSION

The given investigation results demonstrate a close resemblance in annealing behaviour of both intact irradiated and unirradiated CP. Retention of the SiC-layer integrity and diffusion characteristics is an indispensable condition to prevent radioactivity release into the first HTGR circuit and it is provided with stability up to irradiation and annealing temperatures ~2000°C. As a result of first tests possible bursts of neutron flows in the HTGR active zone by 1-2 orders of magnitude don't influence the CP tightness, if the temperature doesn't exceed the critical one.

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DIFFUSION MODELING OF FISSION PRODUCT RELEASE DURING DEPRESSURIZED CORE CONDUCTION COOLDOWN CONDITIONS*

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Abstract

A simple model for diffusion through the silicon carbide layer of TRISO particles is applied to the data for accident condition testing of fuel spheres for the High-Temperature Reactor program of the Federal Republic of Germany (FRG). Categorization of sphere release of ^{137}Cs based on fast neutron fluence permits predictions of release with an accuracy comparable to that of the US/FRG accident condition fuel performance model. Calculations are also performed for ^{85}Kr, ^{90}Sr, and ^{110m}Ag. Diffusion of cesium through SiC suggests that models of fuel failure should consider fuel performance during repeated accident condition thermal cycling. Microstructural considerations in models of fission product release are discussed. The neutron-induced segregation of silicon within the SiC structure is postulated as a mechanism for enhanced fission product release during accident conditions. An oxygen-enhanced SiC decomposition mechanism is also discussed.

1. INTRODUCTION

This report represents a summary of current activities at ORNL related to generic MHTGR fuel performance accident condition (AC) modeling. Work is ongoing in the analysis of existing fuel performance AC codes and models, and determination of relevant phenomena for further model development. Analysis of the recent FRG sphere release data is emphasized because of the minimal particle design variability compared to earlier US and FRG irradiation tests.

Accident condition modeling over the last decade has often emphasized a concept of SiC failure under irradiation and heating rather than a more mechanistic explanation of SiC evolution. A recent model[1] concluded that the fast neutron fluence has little effect on fission product (FP) release which, if true, would make SiC remarkably neutron-resistant. This model also assumed that diffusion of cesium across the SiC layer can be neglected at temperatures above 1600°C in favor of a less-well-defined concept of SiC failure. To test these assumptions, a simple diffusion-based model has been applied to the heating test data for FP release from FRG spheres. This model is based on an analytical expression for diffusion through a thin spherical shell (i.e., the SiC barrier layer). By grouping the sphere release data according to fast fluence, and numerically averaging the diffusion coefficients within each group, predictive capabilities for ^{137}Cs release comparable to the 1989 US/FRG AC model[1] are possible. A general discussion of microstructural phenomena which may be relevant to FP release through SiC is also presented, and some simple conceptual models for enhanced FP release due to SiC damage and degradation are discussed.

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