FISSION PRODUCT BEHAVIOUR IN THE PRIMARY CIRCUIT OF AN HTR

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Introduction

The knowledge of fission product behaviour in the primary circuit of a High Temperature Reactor (HTR) is an essential requirement for the estimations of the availability of the reactor plant in normal operation, of the hazards to personnel during inspection and repair and of the potential danger to the environment from severe accidents.

On the basis of the theoretical and experimental results obtained at the "Institute for Reactor Components" of the KFA Jülich /1/, /2/ the transport- and deposition behaviour of the fission- and activation products in the primary circuit of the PNP-500 reference plant has been investigated thoroughly.

Specially work had been done to quantify the uncertainties of the investigations and to calculate or estimate the dose rate level at different components of the primary cooling circuit. The contamination and the dose rate level in the inspection gap in the reactor pressure vessel is discussed in detail.

For these investigations in particular the surface structure and the composition of the material, the chemical state of the fission products in the cooling gas, the composition of the cooling gas and the influence of dust on the transport- and deposition behaviour of the fission products have been taken into account. The investigations have been limited to the nuclides Ag-110 m; Cs-134 and Cs-137.

1. Description of the reference plant

The PNP-500 plant is planed as a demonstration plant for nuclear process heat application.

The main characteristics of this plant are:

- pebble bed core with OTTO-fuel management;
- He-heating temperature intervall 290 °C - 950 °C;
- mean power density 4.0 MW/m^3;
- integrated system in a prestressed concrete vessel;
- three main cooling loops. One for hydro-coal-gasification (HKV), two for coal gasification with water vapour (WKV-loop);
- He-mass flow: HKV-loop 73.8 kg/sec and WKV-loop 36.9 kg/sec each;
- He-pressure: 40 bar.

In Fig. 1 the primary circuit of the reference plant is shown schematically. The arrows are giving the direction of flow during normal operation. The He leaves the core (1) at a temperature of 950 °C passes through the bottom reflector (2) to the HKV- and WKV-loops. In the HKV-loop the He flows through the hot pipe (3), the reversed flow section (4) to the steam reformer at a temperature of ca. 700 °C, through a gas pipe (6) to the steam superheater (7), the evaporizer (8) and the economizer (9). From here the He flows at 290 °C through a gas pipe (10) to the blower (11) and through another gas pipe (12) back to the outside of the economizer. The He is passing the outside of the steam generator (13), a gas pipe (14), the outside of the steam reformer (15) and the cold gas pipe (16) to the annulus space. Here the gas is mixed with the gas from the other loops. 30 % of the gas flows through the inspection gap and 70 % through the cooling gap between the side reflector and the thermal shield. Finally the gas is led back to the core through the top reflector. In the WKV-loops the He flows

through the hot pipe (3) and the reversed flow section to the He-He-heat exchanger (5), the outside of the heat exchanger (6), a gas pipe (7) and to the blower (8). Through a cold gas pipe (9) the gas is passing also to the annulus space.

The following materials are foreseen for the different components:

- carbon stone for the hot pipe insulation;
- Incoloy 802 or In-617 for the steam reformer and the He-He-heat exchanger repectively;
- Incoloy 800 for the steam generator;
- for the low temperature components ferritic material will be used, for example 15 Mo 3;
- the material for the blower has not been selected yet.

2. Fission product release from the core

For the fission product release from the core the reference values for BISO-particles, highly enriched have been selected. They are / 3 /

Cs - 137: $\dot{R}$ = 150 Ci/a
Cs - 134: $\dot{R}$ = 120 Ci/a
Ag - 110 m: $\dot{R}$ = 500 Ci/a

In general, but especially in case of reducing atmosphere over the operation time of 30 years condensations effects cannot be excluded for Cs and Ag. This means one has also to know the total partial pressure for Cs and Ag. Based on estimations for the total partial pressures for Cs three times the partial pressure of Cs-137 and for Ag, 100 times He partial pressure of Ag-110 m were used for the calculations. Ag-107 gives the main contribution.

3. Basic assumptions

The investigations have been carried out on the basis of the physical model developed by Iniotakis and the computer programmes "PATRAS" and "PATRAS-S". The model gives a phenomenological description of the transport and plate-out of the fission- and activation products /4/, /5/, /6/.

The transport and plate-out behaviour of the fission products depends very much on the flow and temperature conditions in the primary circuit and the manner of their interaction with the component materials. Specially from:

- the chemical state of the fission products;
- the composition of the cooling gas;
- the structure and composition of the surface of the materials and the materials themselves;
- the growing velocity of oxide layers;
- the possible evaporation of the materials of high temperatures.

In addition the presence of dust in the primary circuit plays an important role.

The parameter which have to be used for the calculations can be classified as follows:

1. Operation parameters
2. Fission product specific parameters
   a) for the transport in the gas phase
   b) for the interaction with the surfaces
   c) for the transport (diffusion) into the material
   d) for the interaction with the dust
3. Dust specific parameters
   a) for the transport in the gas phase
   b) for the interaction with the surfaces

Based on the results of our own experiments, using the reported results from literature and specially the specific literature on dust, we did find our model and the computer programmes justified. Furthermore we could evaluate most of the parameters with a sufficient accuracy.

For some of the parameters which are important for the calculation it is not possible today and may be also in the future to specify them sufficiently for a plant in praxis, that is the structure and composition of the surface layers of the materials and the grain size distribution and amount of dust. In addition the solubility ϕ_∞ (the saturation concentration of the fission products in the bulk of the material in [atoms/cm^3]) could not be evaluated with a sufficient accuracy up to now.

To quantify the uncertainties of the calculations resulting from the lack of knowledge of these data sensitivity analysis had been carried out, where in some cases the variation of the parameters had been extended even to values outside the expected areas.

For the variation limits the following assumption had been made:

- The roughness coefficient (the ratio of the real to the geometrical surface area) had been varied between K = 10 - 1 000. For realistic industrial products one should expect K = 50 - 100 under non-oxidizing atmospheric conditions and K = 100 - 1 000 for oxidizing conditions.
- The adhesional distance y_o, which corresponds referring to /4/, /5/, /6/ to the height of the micro roughness had been varied between y_o = 50 - 1 000 Å. Here mono dispersity was assumed in spite of the fact that in reality there is a statistical distribution.
- For the solubility the following values had been used.

$$\phi_{\infty 1} = 1.6 \cdot 10^{22} \cdot e^{-\frac{15\,000}{RT}}$$

$$\phi_{\infty 2} = 8 \cdot 10^{22} \cdot e^{-\frac{15\,000}{RT}}$$

$$\phi_{\infty 3} = 8 \cdot 10^{22} \cdot e^{-\frac{7\,000}{RT}}$$

- Concerning the grain size of the dust the partical radius had been varied between r_p = 0.01 - 10 µm. Also mono dispersity had been assumed.
- The amount of dust had been estimated to be 900 kg in 30 years of operation, taking the mean value of 30 kg/a. Both cases with and without dust had been envisaged.
- Concerning the composition of the cooling gas the two cases non-oxidizing and oxidizing atmosphere had been taken. In the case of the oxidzing atmosphere a pre-oxidation of the component surfaces had been assumed.

4. Results of the sensitivity analysis

All the reported results are from calculations of the HKV-loop.

In Fig. 2 the influence of the roughness coefficient is shown. An operation time of 30 years, an oxidizing cooling gas and no dust had been assumed. On the ordinate the Cs-activity in µCi/cm^2 is given. On the abszissa the position along the HKV-loop is marked starting with the hot gas pipe (3) and ending with the annulus space (17). The numbers referring to the description of the circuit in chapter 1. In addition the surface temperature along the loop is given in the diagram.

One can see from Fig. 2 that the influence of the roughness coefficient is decreasing with increasing temperature and increasing value of the roughness coefficient. At high values of temperature and roughness the deposition of Cs tends to be independent from these parameters.

This result is plausible for Cs. At small values of roughness and low temperatures the degree of coverage tends to be high. At a given limit the cohesional forces cannot be neglected compared to the adhesional forces, condensation effects occur and influencing the desorption kinetic.

For Ag the roughness coefficient does not influence the deposition because the cohesion energy of Ag exceeds the adhesion energy with the surface material. This result is not shown in the Fig. It may be only mentioned here.

In Fig. 3 the influence of the solubility on the plate-out behaviour of Cs-137 is demonstrated. Again an operation time of 30 years, oxidizing gas atmosphere and no dust is assumed. As one can read from these diagram the influence of the solubility is decreasing with increasing temperature and increasing values of solubility. Again this result seems to be plausible because the increase of the diffusion constant and solubility with increasing temperature prevents local saturation can occur.

In Fig. 4 and 5 the influences of the composition of the cooling gas on the deposition behaviour of Cs-137 and Ag-110 m is shown. 30 years operation time and no dust had been assumed.

The influence of the gas atmosphere decreases with temperature. This can be understood by the following considerations. The oxidized layers on the surface of the material which exist in oxidizing atmosphere serve as a diffusion barrier for Cs. The diffusion constant of Cs in oxide is several orders of magnitude smaller compare to that in the metal. This is resulting in the case of more oxidizing atmosphere in spite of the faster diffusion processes in prevention of local saturations. This is also the fact at high temperatures in the case of oxidizing atmosphere.

In the case of Ag-110 m one can read from Fig. 5 that the influence of the atmospheric conditions is most important for the economizer.

In Fig. 6 the influence of the dust is shown. Curve 1 referes to the dust free system. Curve 2 gives the deposition of Cs-137 on the surfaces assuming there is dust in the system. Curve 3 represents the amount of Cs-137 which is sticking on the dust particles which are themselves be deposited on the surfaces. In this case an adhesional distance $y_o = 300$ Å and a particle radius of $r_p = 1$ µm had been chosen.

One can see in Fig. 6 that there is a considerable influence of the dust on the plate-out.

The shown diagrams, Fig. 2 - Fig. 6, are examples demonstrating typical results from the extended investigations. Within the scope of this paper it is not possible to present all the results from this work in detail. But some additional general results should be mentioned in qualitative form:

- The dust has the strongest influence on the plate-out behaviour compared with the other parameters.
- In general the presence of dust in the system reduces the influences of the other parameters.
- The reason for this is the considerable reduction of the concentration of free fission product atoms in the gas phase. Condensation effects in the cold part of the circuit are reduced or eliminated.
- The influence of dust can be neglected if the particle radius exceeds $r_p \geq 5$ µm.

- Neglecting the dust leads in general to pessimistic results with respect to contamination of the inspection gap and the blowers.

- In the WKV-loops the influence of the roughness, the solubility and the composition of the gas is very small or negligible depending on the special design of the heat exchanger.

5. Estimation of the maximum level of contamination

To estimate the maximum level of contamination four different cases, reference cases, have been calculated. The contamination and dose rate in the inspection gap is used as an example. The reference cases have been chosen in a manner that the influence of the different parameters and the influence of the materials are shown on the one side, on the other side the maximum level of contamination can be estimated. To be in the pessimistic side the influence of the dust is neglected in these calculations.

The reference cases are defined:

Reference case 1

The material of the steam reformer, the steam generator and the heat exchanger is Incoloy-800; the other components are made out of 15 Mo 3 with the exception of the blower. There is oxidizing gas atmosphere and the surfaces of the components are preoxidized.

Reference case 2

As case 1, but In-617 as the material for the steam reformer, the steam generator and the heat exchanger.

Reference case 3

As case 1, but non-oxidizing atmosphere.

Reference case 4

As case 2, but non-oxidizing atmosphere.

In Fig. 7, Tab. 1, the influence of the solubility, roughness for the different cases is given. One can read from this table that within the expected area for the roughness coefficient K = 50 - 100 for the cases 3 and 4 and K = 100 - 1 000 for the cases 1 and 2 all values are within a factor of two. The expected value for the solubility for Cs is about equal to $\phi_{\infty 2}$.

In Fig. 8, Tab. 2, the expected contamination of the inspection gap is given. The contribution of Cs-137, Cs-134 and Ag-110 m for the different cases are shown. The expected values are based on the assumption tha the value for the solubility of Ag is equal to $\phi_{\infty 3}$. For the roughness coefficient K = 50 for non-oxidizing atmosphere and K = 100 for oxidizing atmosphere was assumed. As one can see the main contribution for the contamination originates from the Ag.

In Fig. 8, Tab. 3. the corresponding dose rates are given which are calculated for the center of the inspection gas. Again the Ag is dominating.

These values are pessimistic because the influence of the dust has been neglected.

Summary

The accuracy calculating the distribution of fission products which are deposited on the surfaces in a primary cooling gas circuit of a high temperature reactor depends on the knowledge of the value of a large number of parameters. Most of the parameters estimate by a large

number of experiments in the meantime. For some of the parameters the knowledge is poor (solubility) or they cannot be defined well enough for a practical plant (roughness, composition of the surface, grain size and amount of dust). To get a better knowledge of the uncertainties resulting from the uncertainties of these parameters, sensitivity stuides have been made and demonstrated in some examples within this paper.

Furthermore the maximum value of the contamination and dose rate level in the inspection gap of a PNP-500 Prototype plant has been estimated.

References

/1/ N. Iniotakis, J. Malinowski, K.H. Münchow
Initial results of investigations into fission products deposition in in-pile experiments
Nuclear Engineering and Design 34 (1975), 169-180

/2/ N. Iniotakis, J. Malinowski, H. Gottaut, K.H. Münchow
Results from plate-out investigations
IAEA-meeting, Jülich, December 2.-4., 1980

/3/ K. Röllig
private communication

/4/ N. Iniotakis
Dissertation (not published)

/5/ N. Iniotakis, C.-B. von der Decken
The influence of dust on the behaviour of fission products in high temperature reactors
ENS/ANS Topical Meeting on Nuclear Power Reactor Safety, Brüssel, 16.-19.10.1978
Proceedings, Vol. 3, 2336 - 2347

/6/ C.-B. von der Decken, N. Iniotakis, F.P.O. Ashworth
A Model for the Description of Fission Product Behaviour in the Primary Circuit of a High Temperature Reactor
Conference on Gas Chemistry in Nuclear Reactors and Large Industrial Plant,
Salford, UK, April 1980, Preprints

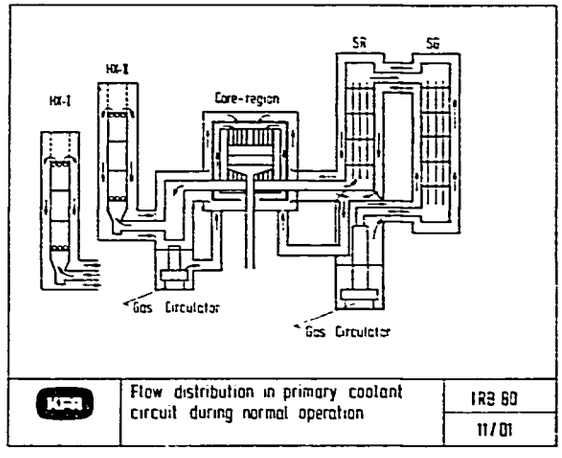

Fig. 1

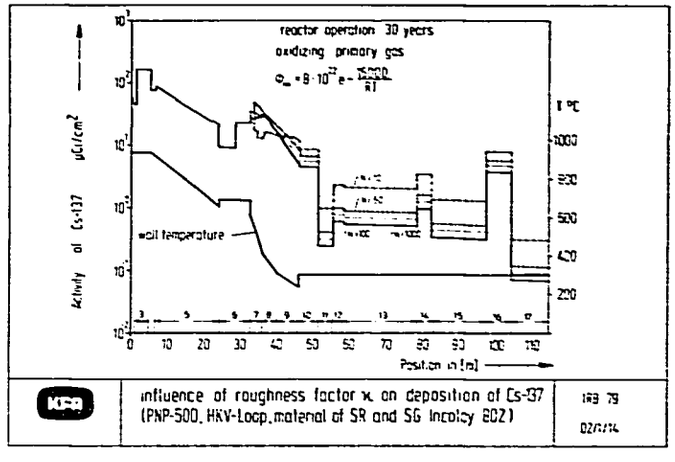

Fig. 2

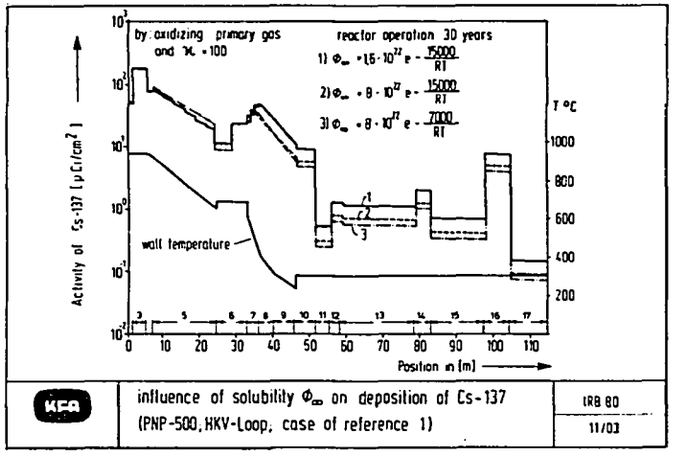

Fig. 3

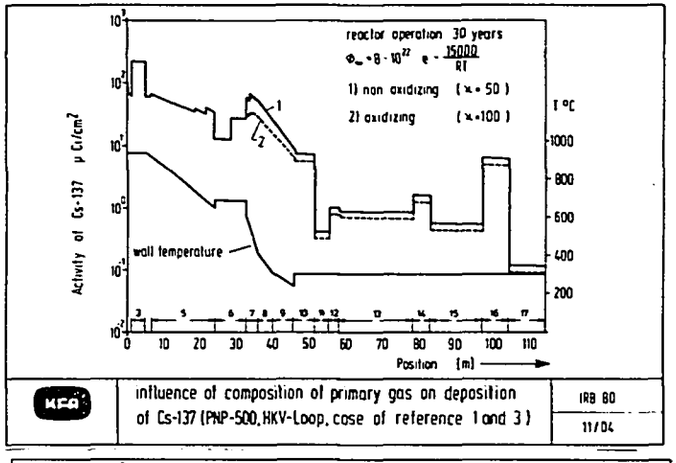

Fig. 4

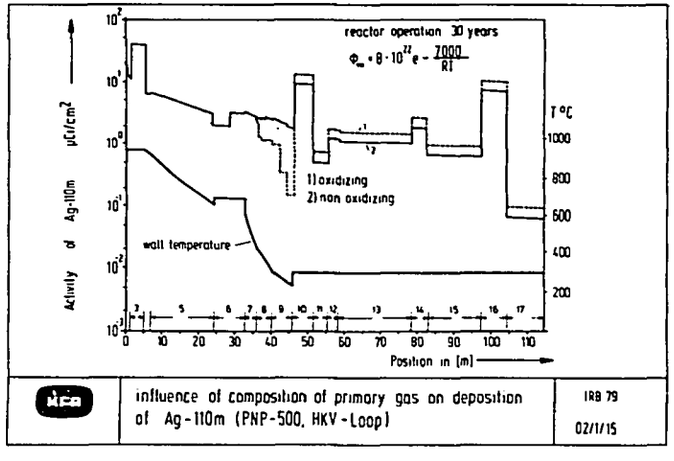

Fig. 5

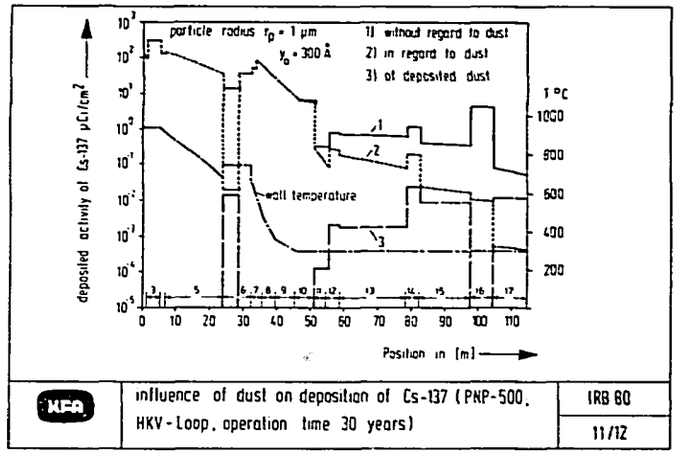

Fig. 6

TABLE 1: INFLUENCE OF SOLUBILITY Φ_∞, ROUGHNESS FACTOR K AND MATERIAL ON Cs-137-CONTAMINATION OF THE INSPECTION GAP; CONTRIBUTION OF THE HVK-LOOP IN Ci AFTER 30 YEARS.

	Case 1			Case 3			Case 2		Case 4	
K	$\Phi_{\infty 1}$	$\Phi_{\infty 2}$	$\Phi_{\infty 3}$	$\Phi_{\infty 1}$	$\Phi_{\infty 2}$	$\Phi_{\infty 3}$	$\Phi_{\infty 2}$	$\Phi_{\infty 3}$	$\Phi_{\infty 2}$	$\Phi_{\infty 3}$
1	1.88	1.00	0.48	0.77	0.63	0.39	1.34	0.50	0.84	0.46
5	0.68	0.38	0.27	0.33	0.29	0.26	0.59	0.27	0.41	0.33
100	0.49	0.30	0.24	0.27	0.25	0.24	0.46	0.24	0.34	0.30
1000	0.32	0.23	0.21	0.22	0.21	0.21	0.34	0.21	0.27	0.21

HINTS: RANGE EXPECTED: K = 100 - 1000 ABOUT AND $\Phi_{\infty 2}$ NEARLY
REFERENCE CASES:
CASE 1 AND 2 FOR K = 100 AND $\Phi_{\infty 2}$
CASE 3 AND 4 FOR K = 50 AND $\Phi_{\infty 2}$

Fig. 7

TABLE 2: CONTAMINATION OF THE INSPECTION GAP AFTER 30 YEARS IN Ci

Case	Cs-137	Cs-134	Ag-110m
1	0.44	0.05	0.79
2	0.62	0.08	0.79
3	0.37	0.05	0.57
4	0.52	0.08	0.57

TABLE 3: DOSE RATE IN THE INSPECTION GAP AFTER 30 YEARS IN MREM/H

Case	Cs-137	Cs-134	Ag-110m	TOTAL
1	6	3	47	56
2	9	5	47	61
3	5	3	34	42
4	7	5	34	46

Fig. 8