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Investigation of NACOK Air Ingress Experiment Using Different System Analysis Codes

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Abstract – *Air ingress into to the core after the primary circuit depressurization due to large breaks of the pressure boundary is considered as one of the severe hypothetical accidents for the High Temperature gas-cooled Reactor (HTR). If the air source and the natural convection cannot be impeded, the continuous graphite oxidation reaction along with the formation of burnable gas mixtures resulting in the corrosion of the fuel elements and the reflectors might damage the reactor structure integrity and endanger the reactor safety. In order to study the effects of air flow driven by natural convection as well as to investigate the corrosion of graphite, the NACOK (Naturzug im Core mit Korrosion) facility was built at Jülich Research Center in Germany. A complete 2A-rupture of the coaxial duct in the HTR primary system, as well as the chimney effect caused by breaks in both upper and lower parts of the pressure boundary was simulated in the test facility. Several series of experiments and the related code validations (TINTE, DIREKT, THERMIX/REACT, etc.) have been performed on this facility since the 1990s. In this paper, the latest NACOK air ingress experiment, carried out on October 23, 2008 to simulate the chimney effect, was preliminary analyzed at NRG with the SPECTRA code, as well as at INET, Tsinghua University of China with the TINTE and THERMIX/REACT codes. The calculating results of air flow rate of natural convection, time-dependent graphite corrosion, and temperature distribution are compared with the NACOK test results. The preliminary code-to-experiment and code-to-code validation successfully proves the code capability to simulate and predict the air-ingress accident. Besides, more research work, including parameter sensitivity analysis, modeling refinement, code amelioration, et al, should be performed to improve the simulation accuracy in the future.*

Keywords – *High temperature gas-cooled reactor, air ingress, graphite corrosion, NACOK test, code validation*

I. INTRODUCTION

Air ingress accident, which will result in graphite oxidation reaction of fuel elements and reflectors so as to weaken the structural strength, damage the graphite structure integrity of the core bottom, impact retention capacity of coated particle and produce the flammable gas mixtures, is considered as one of the severe hypothetical accidents for the High Temperature gas-cooled Reactor (HTR).

The heterogeneous chemical reactions between graphite and oxygen include:

Complete oxidation

$C + O_2 \rightarrow CO_2$ ΔH=-3.94×10^5 J/mol

Incomplete oxidation

$2C + O_2 \rightarrow 2CO$ ΔH=-2.22×10^5 J/mol

Boudouard reaction

$C + CO_2 \leftrightarrows 2CO$ $\Delta H=1.71\times10^5$ J/mol

The main homogenous reaction referring to the air ingress accident is:

$2CO + O_2 \rightarrow 2CO_2$ $\Delta H=-5.64\times10^5$ J/mol

The mass transfer and diffusion play important role in the reaction rate between the gas and the porous graphite. Accordingly, the chemical reaction could be divided into the following three types:

I *CR (chemical regime) at low temperature*

The reaction rate is very slow, the gas transfer process in the graphite can be neglected and the reaction takes place in total graphite homogeneously.

II *IPDR (in-pore diffusion controlled regime) at middle temperature*

The gas transport process cannot be neglected. The porous diffusion and chemical reaction both determine the gas convention rate.

III *BLDR (boundary layer diffusion controlled regime) at high temperature*

The chemical reaction takes place at the graphite boundary and the boundary layer diffusion determine the corrosion rate.

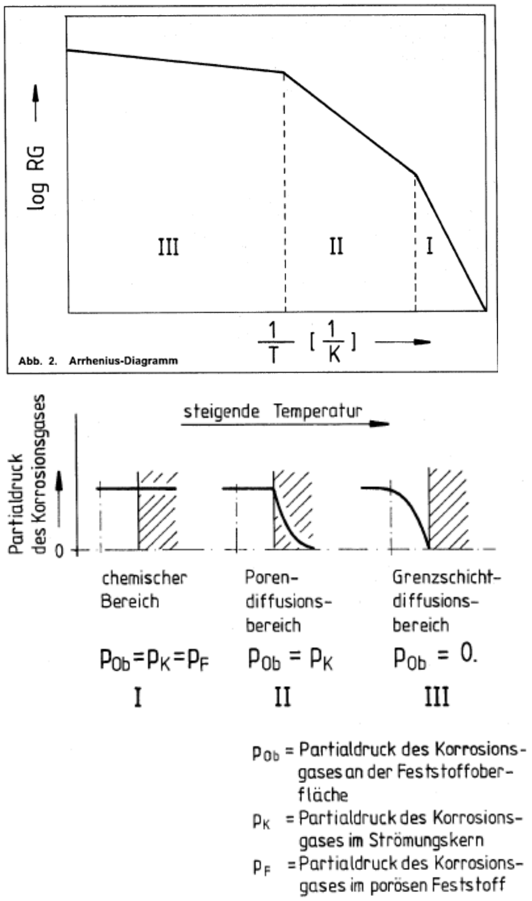


Fig. 1: Graphite/gas reaction characteristics.

The temperature range for above three reaction types are approximately as follows:

CR: <500°C

IPDR: 500°C - 900°C

BLDR: >900°C

The graphite/gas reaction characteristics are described as shown in Fig. 1.

The NACOK (Naturzug im Core mit Korrosion) facility was built at Jülich Research Center in Germany to study the oxidation behaviour of graphite blocks with air flow driven by natural convection (chimney effect) in the event of a complete rupture of the coaxial duct or a break of larger tubes in the HTR primary system.

In this paper, a preliminary analysis is performed to investigate the newly NACOK air ingress experiment carried out on October 23, 2008, which simulates the chimney effect and adopts different block geometries with two different graphite grades. A detailed simulation model of this NACOK facility was developed at NRG with the SPECTRA code and at INET, Tsinghua University of China, with the TINTE and THERMIX/REACT codes. The calculations were performed to validate those codes for the HTR air ingress scenarios.

II. NACOK FACILITY

The overall experimental arrangement of the NACOK facility is shown in Fig. 2. The inner diameter of the inlet tube is 0.125m. During the chimney test, the return tube is unavailable, and the air can flow out from the top of the experimental channel. The experimental channel, with a cross section of 30 × 30 cm and a height of more than 8 meters, can be heated up to 1200°C by electric heaters with a maximum power of about 150 kW.

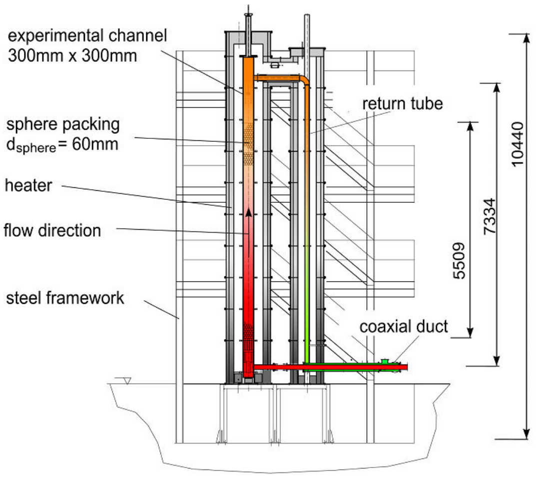
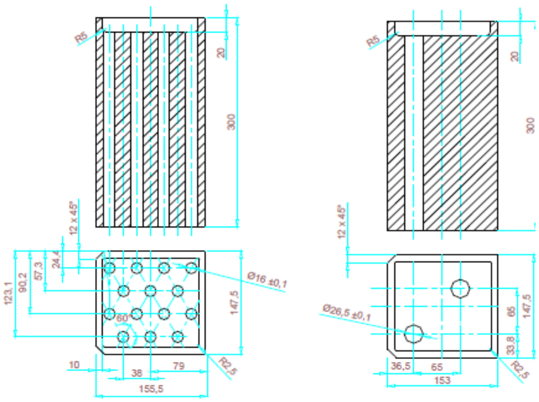
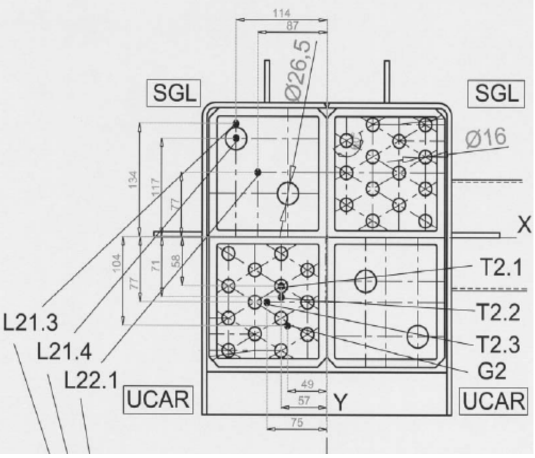


Fig. 2: NACOK test facility.

To provide an appropriate pressure drop, a pebble bed of 0.3 m high, filled with small (10 mm) ceramic spheres, is placed in the upper part of the main channel. In the lower part of the main channel, three levels of graphite blocks are installed, as shown in Fig. 3.



3a: Geometric types of test specimens



3b: Arrangement of graphite blocks

Fig. 3: Graphite block configuration of test.

Each graphite block is 0.3 m high. Two types of geometry of the graphite blocks have been selected to study their impact under comparable conditions: one has 14 holes of 16 mm in diameter, while the other has 2 holes of 36.5 mm in diameter.

Besides, two different graphite types are used to make each of the above mentioned geometries: Graphite UCAR PCEA and Graphite SGL NBG17.

The following test procedure was applied:

1) The test facility was uniformly heated by blowing nitrogen at 900˚C (1173 K) into the facility for a sufficiently long time to ensure that all components are at thermal equilibrium.

2) When this equilibrium had been achieved the entrance duct was opened and air from the building was allowed to enter with temperature of 20˚C (293 K) and relative humidity (RH) of 0%.

3) The inlet valve position was set to allow the air flow of about 5×10^{-3} kg/s. The valve resistance (referring to the flow area of a D125 mm pipe) was about 110;

4) The steel walls of the main channel were kept at the desired temperature by the external heaters.

There are two main flow resistances for this test, the pebble bed and the inlet valve. The resistance introduced by the graphite blocks is minor, compared to the other two resistances.

Many measuring points are set in the facility, including the temperature, gas composition, et, al. The experimental results indicate that the main reaction product is CO_2, which means the complete oxidation occurring at experimental temperature.

III. SPECTRA SIMULATION

III.A. Code Introduction

The SPECTRA (Sophisticated Plant Evaluation Code for Thermal-hydraulic Response Assessment) code, developed at NRG, the Netherlands, is a computer program designed for thermal-hydraulic analyses of nuclear or conventional power plants.

In the past the code has been used for HTR analyses, including independent verification of the thermal-hydraulic analyses of PBMR, dust transport, deposition, and resuspension calculations in the PBMR and NGNP, as well as the Reactor Cavity Cooling System analysis of PBMR.

In the future an air ingress and water ingress analyses of HTR are foreseen. For this reason the code is being validated for the air and water ingress scenarios.

III.B. Graphite Oxidation Model

SPECTRA uses a general formula, applicable for oxidation of a variety of materials. The reaction kinetics coefficients must be supplied by the user, or taken from the built-in database containing values for zircaloy, steel, and graphite.

For oxygen reaction with graphite $\mathbf{C+O_2 \rightarrow CO_2}$, three alternative models have been applied:

No, Kim, Lim (NKL) correlation [5]:

$$R = 7500 \cdot \exp\left(-26218/T\right) \cdot P_{O2}^{0.75}$$

R: reaction rate [kg/m^2/s] (kg of oxidized C)
T: temperature [K]
P_{O2}: oxygen partial pressure [Pa]

CEA correlations [3]:

- graphite UCAR PCEA

$$R = 25.6 \cdot \exp\left(-18040/T\right) \cdot P_{O2}^{0.50}$$

- graphite SGL NBG 17

$$R = 88.1 \cdot \exp(-19240/T) \cdot P_{O2}^{0.50}$$

There is very small difference between these two correlations - see Fig. 4. The second correlation (SGL NBG 17) was used in SPECTRA calculation.

Roes correlation [6]:

Roes correlation takes into account not only the temperature but also the gas velocity. The correlation has the form:

$$\frac{dm^x}{dt} = \left(\frac{1}{K_T(T)} + \frac{1}{K_v(v,T)} \right)^{-1} \cdot K_p(p)$$

The reaction coefficients are [6]:

- $A = 7.2\text{E+9}$ [mg/cm^2/hr]
- $B = 16{,}140.0$ [K]
- $C = 770.0$ [(mg/cm^2/hr) / (m/s)^D / K^E]
- $D = 0.65$
- $E = 0.34$

Converted to SI units (required by SPECTRA):

- $A = 7.2\text{E+9}$ [mg/cm^2/hr]
- $A = 7500$ [kg/m^2/s]
- $B = 16{,}140.0$ [K]
- $C = 8.03 \times 10^{-4}$ [(kg/m^2/s) / (m/s)^D / K^E]
- $D = 0.65$
- $E = 0.34$

Comparison of the correlations for oxygen reaction is shown in Fig. 4. In case of Roes correlation the gas velocity of 1.0 m/s is used, which is approximately equal to the gas velocity in the NACOK test.

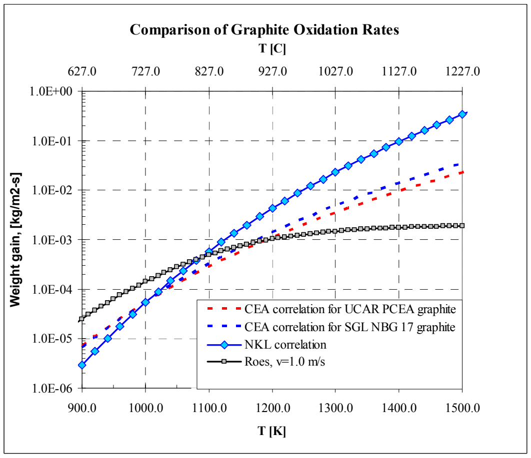


Fig. 4: Different correlations of oxygen reactions.

For CO_2 reaction with graphite $\mathbf{C+CO_2 \rightarrow 2CO}$, Moorman correlation is used:

$$R = \frac{0.145 \cdot \exp(-25000/T) \cdot P_{CO2}}{1 + 3.4 \times 10^{-5} \cdot \exp(7000/T) \cdot P_{CO2}^{0.5}}$$

P_{CO2}: carbon dioxide partial pressure [Pa]

For the purpose of computer code calculations, the following correlation had been developed from Moorman correlation:

$$R = 0.70 \cdot \exp(-26000/T) \cdot P_{CO2}^{0.80}$$

III.C. SPECTRA Model of NACOK

III.C.1. Test Channel

The nodalization applied to model the main channel as well as the inlet and outlet pipes of the NACOK facility is shown in Fig. 5. In order to model pre-conditioning of the test facility, a nitrogen tank and valve (CV-098 and JN-098) are included. The CV-098 is filled with nitrogen at 1173 K).

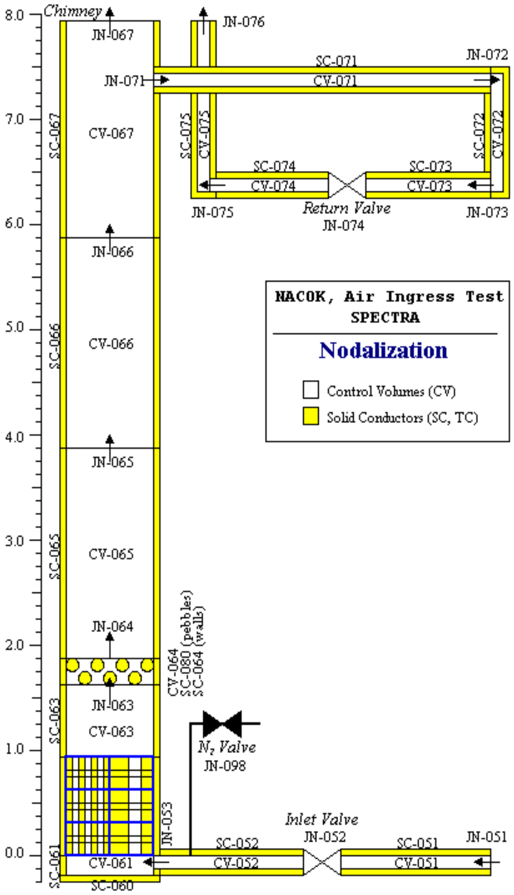


Fig. 5: NACOK model - test channel.

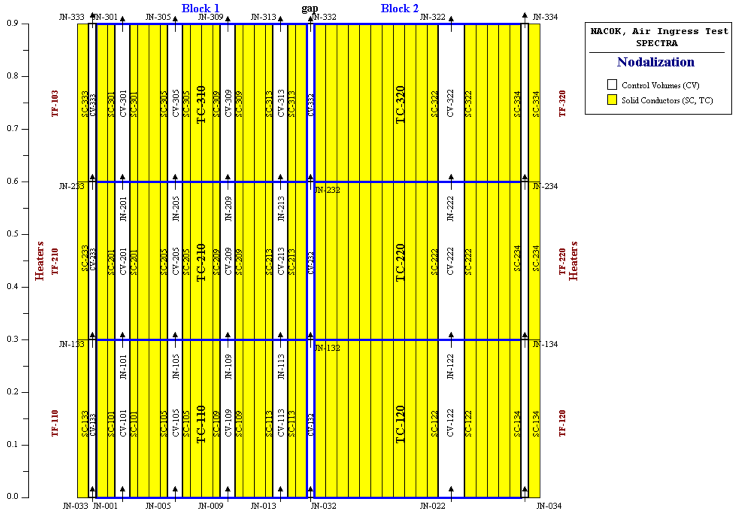


Fig. 6 NACOK model - graphite blocks

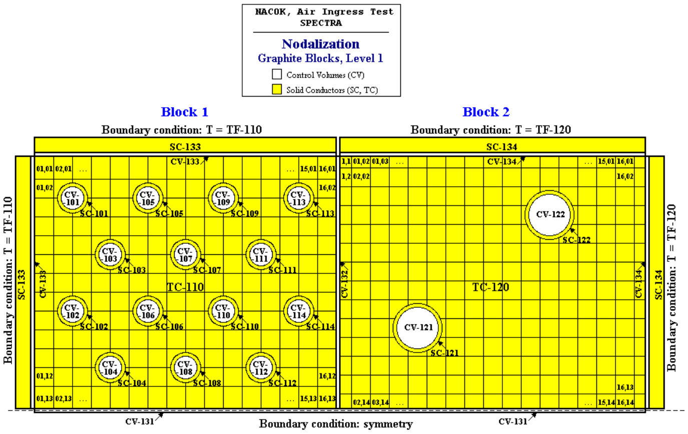


Fig. 7 NACOK model - graphite blocks

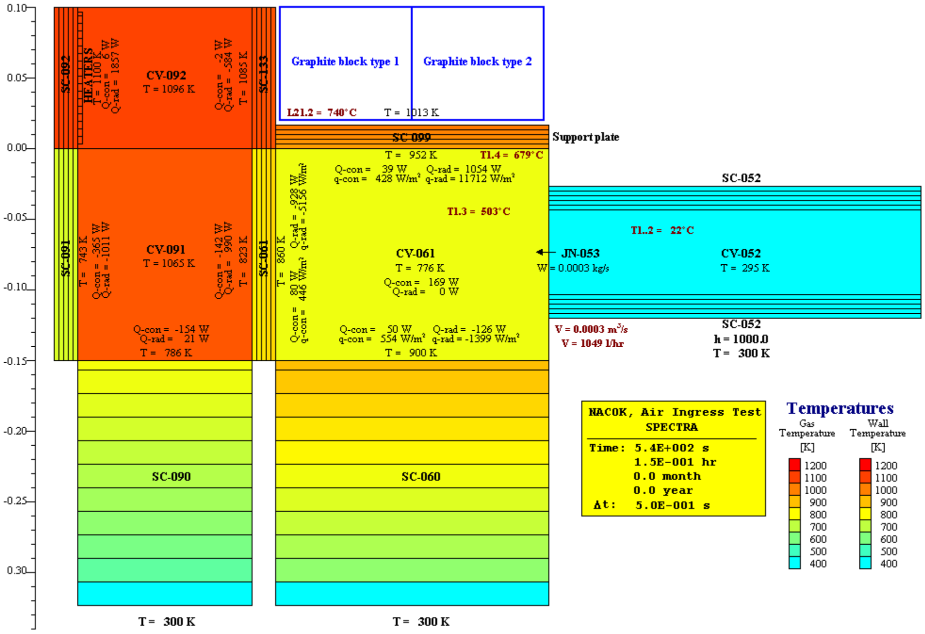


Fig. 8 NACOK model - inlet part - results of the pre-conditioning phase

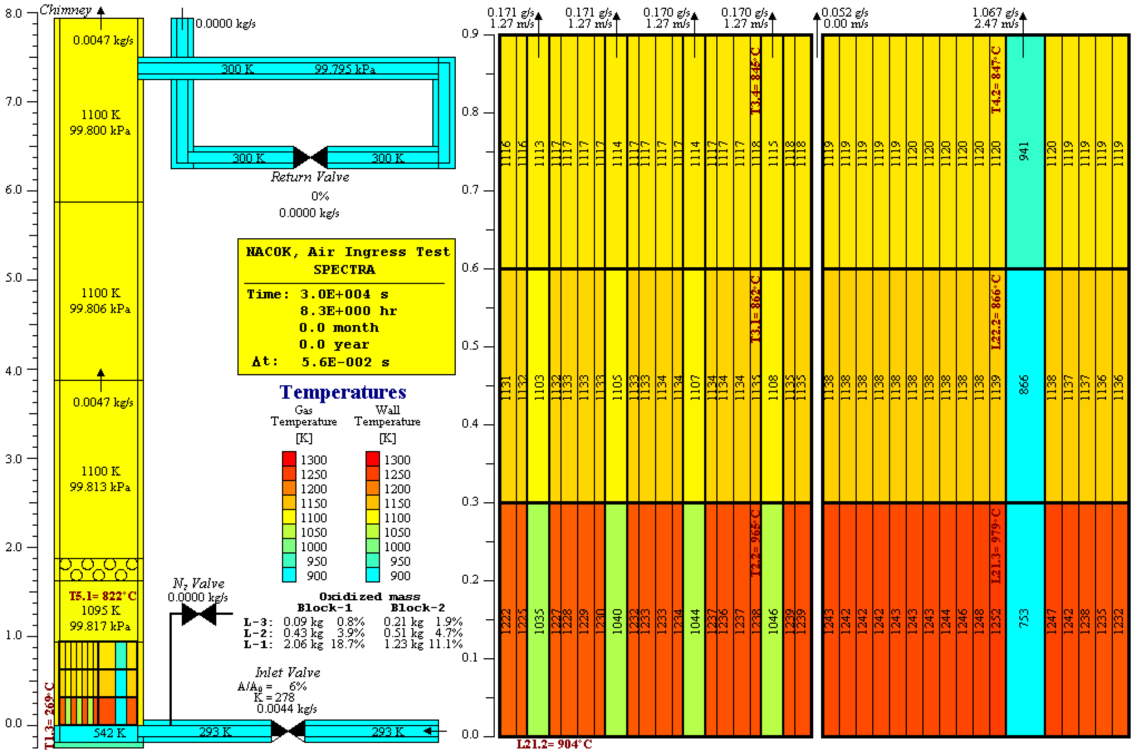


Fig. 9: SPECTRA results, Roes correlation, vertical layout at t = 30,000 s

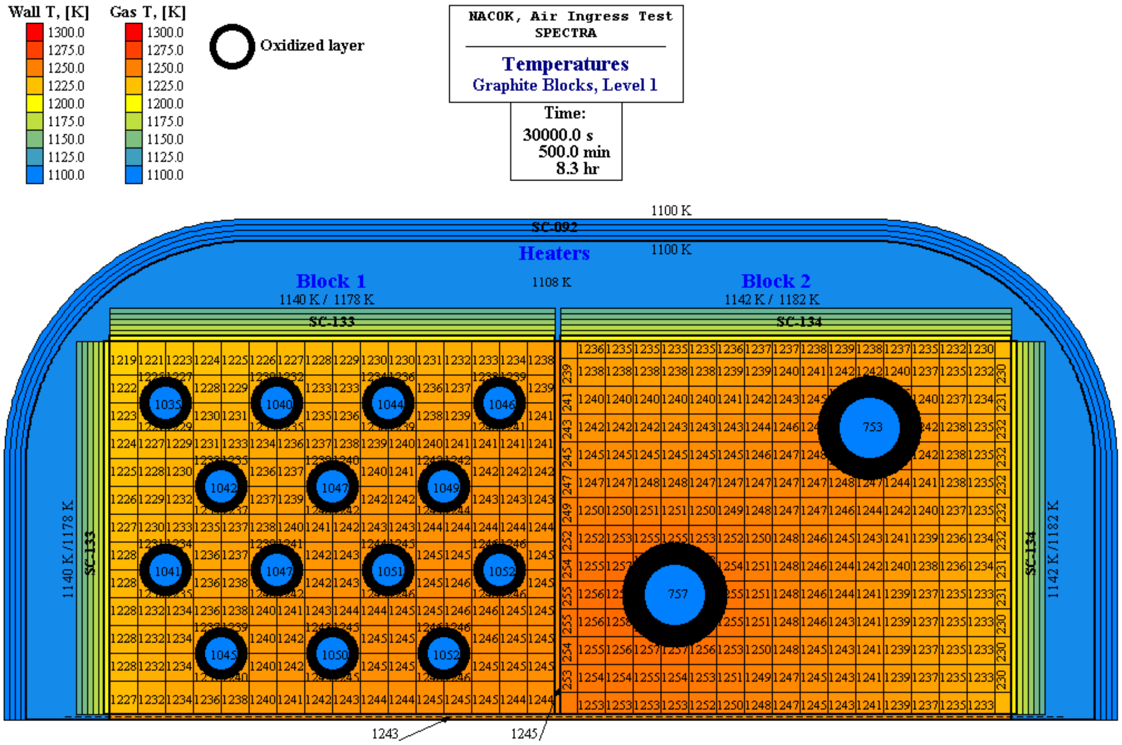


Fig 10: SPECTRA results, Roes correlation, layer 1 (bottom) at t = 30,000 s

IV.C.2. Graphite Blocks

Nodalization of the graphite blocks is shown in Fig. 6 and Fig. 7. The following assumptions are made:

1) The graphite blocks are modelled as a 2-D rectangular geometry structures, conducting heat in the *x-y* directions (horizontal).

2) The difference in the properties of graphite UCAR PCEA and SGL NBG17 are considered to be negligible. Therefore only two blocks are modelled and a symmetry boundary condition is applied.

3) In vertical direction, the blocks are represented by 3 levels, each with height of 0.3 m. The vertical layout is shown in Fig.6.

4) The horizontal, *x-y*, nodalization of the graphite blocks is shown in Fig. 7. To simulate the heaters, the boundary conditions on the outer side of the steel walls are defined by Tabular Functions. The temperature (1100 K) in the heater area was taken from experimental data.

5) Heat loss in vertical direction, from Block 1 through the support structure is taken into account. The representative conductance between the block 1 and the support structure has been found by trial and error to match the initial temperature of the support structure. The value of $h = 400$ W/m^2-K was used. It is quite important to model this heat flow, as will be shown below.

Preliminary test calculations showed that it is very important to correctly model complicated radiative and convective heat transfer in the lower part of the test facility. An accurate temperature distribution during the pre-conditioning phase, and therefore an accurate starting point for the experiment, must be obtained, otherwise comparison of the oxidation model becomes impossible. The results obtained for the pre-conditioning phase are shown in Fig. 8. There is a heat loss through the floor through the insulation and through the lower part of the facility walls. The heaters are providing an approximately constant temperature in the annular space above the test facility.

III.D. Simulation Results

Temperatures of the graphite blocks and the lower ceramic support structure are shown in Fig. 11 through Fig. 14, and Fig. 14. Results closest to experiment are obtained when the velocity-dependent correlation from Roes is used (Fig. 14).

Application of the NKL correlation (which gives the highest reaction rates for T>900°C - Fig. 4) leads to overestimation of the reaction rates in the lower block. Consequently the oxidation of the middle and the top blocks are underestimated (most oxygen is already consumed by the block 1) - Fig. 12.

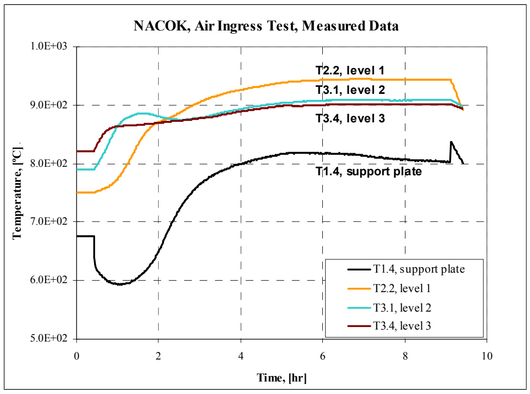


Fig 11. Structure temperatures, experiment

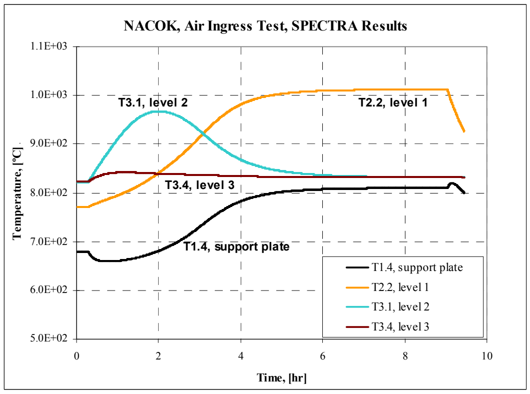


Fig 12. Structure temperatures, SPECTRA, NKL

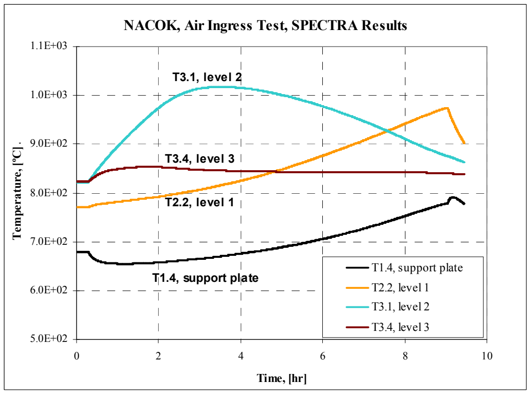


Fig 13. Structure temperatures, SPECTRA, CEA

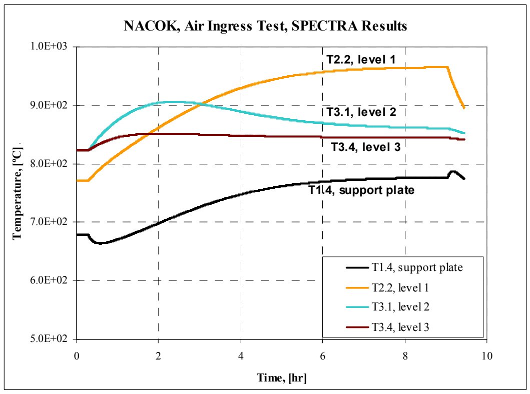


Fig 14. Structure temperatures, SPECTRA, Roes

CEA correlation gives relatively low reaction rates for T~800-850ºC - Fig. 4. This leads to underestimation of reaction rates in the lower block where the cold gas enters. The oxidation reaction produces relatively low amount of heat and the lower block temperature rise is very slow. Consequently a lot of oxidation occurs in the middle block. The lowest block becomes hottest only after about 7.5 hours - Fig. 13, while in thwe experiment this occures already after about 2 hours (Fig. 11).

Summary of the graphite mass loss is given in Table 1. The experimental data obtained for two different kinds of graphite, therefore two numbers are shown. Note that in the SPECTRA model the different grapite types is ignored - section IV.C.2. It is seen that the best agreement with experiment is obtained using the Roes correlation.

Geom		SPECTRA			Data
Type	Layer	NKL	CEA	Roes	[%]
1	bot	24	21	22	23-24
2		20	14	14	11-16
1	mid	1.7	4.3	3.1	3.4-3.5
2		2.4	6.2	3.7	4.5-5.0
1	top	0.3	0.5	0.7	0.8-1.5
2		0.5	1.2	1.8	2.6-2.8

Table 1: Graphite mass lost, SPECTRA

IV. TINTE AND THERMIX/REACT SIMULATION

IV.A. Code Introduction

TINTE code is specifically developed to assess the transient behavior of pebble-bed high temperature gas-cooled reactor by Jülich Research Center of Germany. The thermo-fluid dynamics by different heat transport characteristics and the chemical processes between water, air and graphite on the basis of new experimental results are taken into consideration. The influence of the mass transfer and diffusion is also included. The code has been validated in VELUNA corrosion experiments, as well as the earlier NACOK experiments.

Besides the oxidation reaction introduced in part I, other heterogeneous and homogeneous reactions induced by water ingress are also considered in TINTE code.

For the graphite oxidation, the reaction rate used in TINTE code is derived from experiment:

$$-\dot{q}_{O_2} = \frac{1.5 k_1 R T c_{O_2} F_{b1}}{P_{GES}(1+(k_2 R T c_{O_2})^{0.4})^{1.25}}$$

$$k_1 = 5.83\times10^{4}\exp(-1.1\times10^{5}/RT)[\frac{mol}{m^2 s}]$$

$$k_2 = 5.5\times10^{-14}\exp(1.8\times10^{5}/RT)[\frac{1}{Pa}]$$

$$F_{b1} = (0.447 + 0.8094b - 0.3221b^2 + 0.0681b^3$$
$$- 0.00613b^4 + 1.232\times10^{-5}b^5 + 2.89\times10^{-5}b^6$$
$$- 1.15\times10^{-6}b^7)/0.447$$

P_{GES}: total gas pressure [Pa]
R: 8.31441 [J/mol/K]
T: gas temperature [K]
c_{A}: molar concentration of gas A [mol/m^3]
F_{b1}: corrosion factor
b: graphite corrosion proportion [%]

With lower concentration in gas mixture, the approximative reaction rate of CO and CO$_2$ is:

$$\dot{q} = \beta c_W$$

$$k_{CB} = \frac{[c_{WCO}]^2}{[c_{WCO_2}]}$$

β: mass transfer coefficient [m/s]
c_{W}: molar density on the graphite surface [mol/m^3]

so:

$$\frac{[\dot{q}_{CO}]^2}{\dot{q}_{CO2}} = k_{CB}\frac{\beta_{CO}{}^2}{\beta_{CO2}} = k'_{CB}$$

and:

$$\frac{1}{2}\dot{q}_{CO} + \dot{q}_{CO2} = |\dot{q}_{O2}|$$

$$\frac{\dot{q}_{CO}{}^2}{k'_{CB}} + \frac{1}{2}\dot{q}_{CO} = |\dot{q}_{O2}|$$

So for the above two oxidation:

$$\dot{q}_{CO} = \frac{1}{4}(-k'_{CB} \pm \sqrt{k'^{2}_{CB} + 16k'_{CB}|\dot{q}_{O_2}|})$$

$$\dot{q}_{CO2} = |\dot{q}_{O_2}| + \frac{1}{8}(k'_{CB} \mp \sqrt{k'^{2}_{CB} + 16k'_{CB}|\dot{q}_{O_2}|})$$

For complete oxidation, considering F_{b1} as 1, the reaction rate is shown in Fig. 15.

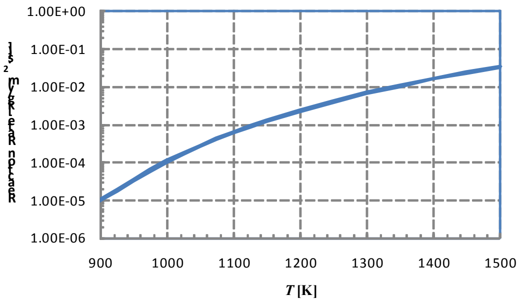


Fig. 15: Reaction rate in TINTE code.

THERMIX/REACT, also developed by Jülich Research Center, is another code used to analyze the water-ingress and air-ingress accidents.

The reaction rate of carbon oxidation in IPDR used in this code is:

$$\dot{r} = \frac{k_{01}\exp(-\frac{E_1}{RT})\cdot {}^{S}P_{ED}\cdot f_1(B)f_2(P_{GES})}{1+k_{02}\exp(-\frac{E_2}{RT})\cdot {}^{S}P_{ED}^{0.5}+k_{03}\exp(-\frac{E_3}{RT})\cdot {}^{S}P_{\mathrm{Pr}od}^{0.5}}$$

k: reaction rate constant

E: activity energy [J/mol]

${}^{s}P_{\mathrm{ED}}$, ${}^{s}P_{\mathrm{Prod}}$: the partial pressure of the reaction gas and production gas at the graphite surface [Pa]

f_1, f_2: function describing the pressure and corrosion-dependent reaction rate.

While in BLDR:

$$-\frac{1}{F}\frac{dn_{ED}}{dt} = \frac{\beta}{RT}({}^{H}P_{ED} - {}^{S}P_{ED})$$

F: surface area [m^2]

n_{ED}: molar number [mol]

${}^{\mathrm{H}}P_{\mathrm{ED}}$: the partial pressure of the reaction gas at the main flow [Pa]

	k_{01}	k_{02}	k_{03}	E_1/R	E_2/R	E_3/R
Pebble bed	0.121	1.43e-2	0	9768	-141	0
Reflector	0.476	0	0	12000	0	0

Table 2: Reaction factors in THERMIX/REACT .

Table 2 lists a group of experimental data of activity energy and reaction rate constant derived from experiments. For lake of sufficient experimental data of the graphite type used in NACOK 2008 test, above data are utilized at present in THERMIX/REACT model.

Considering the total pressure as 1.013×10^5 Pa, O_2 partial pressure of 0.22×10^5 Pa, and ignoring influence of the reaction production partial pressure and graphite corrosion, reaction rate of complete oxidation is shown in Fig. 16.

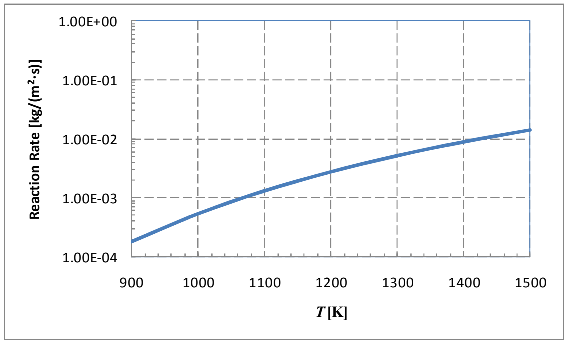


Fig. 16: Reaction rate in THERMIX/REACT code

IV.B. Calculating Model

Fig. 17 shows the calculating model for TINTE code and THERMIX/REACT code according to the NACOK experiment facility. The rectangle experimental channel is modeled as a circular tube with the same equivalent diameter. TINTE and THERMIX/REACT adopt a two-dimensional r-z geometry model, so the arrangement of those two type graphite blocks could not be simulated accurately. Therefore, a kind of homogeneous graphite with certain porosity is considered in this model.

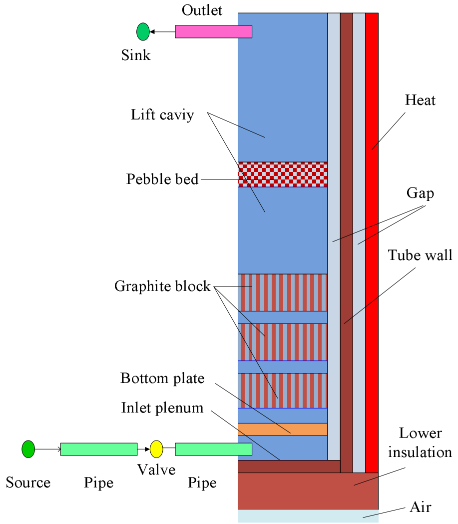


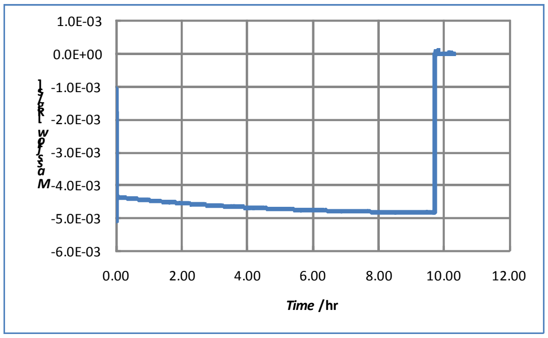
Fig. 17: Calculating model for TINTE and THERMIX/REACT

IV.C. Simulation Results

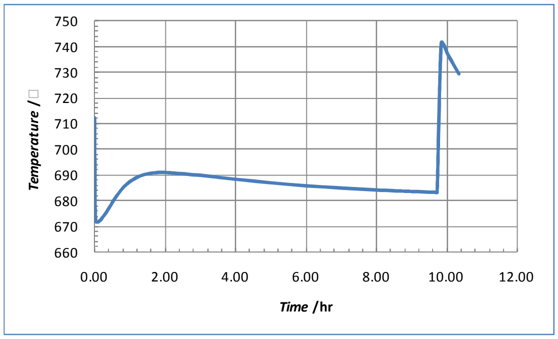
Fig. 18 indicates the TINTE calculating results of mass flow and gas temperature in the inlet plenum. During the test, the average air mass flow is approximately 4.32 g/s, and the experimental result of gas temperature in the inlet plenum is shown in Fig. 19 (the lowest curve).

The lift force, caused by the density difference between the hot gas and the cold gas, results in the natural convection of the air. The calculating result of the mass flow accords well with the experimental one.

If the valve is opened, the cold air will flow into the experimental channel from the inlet tube, so the gas temperature at the inlet plenum will drop down quickly too. However, due to the heaters keeping the channel wall with a constant temperature of 900°C, as well as the graphite oxidation releasing a great deal of heat, the graphite will maintain high temperature. The gas temperature could be expected to increase again after a short-time decrease for the hot graphite upwards and the heat radiation.



18a air mass flow



18b temperature in the inlet plenum

Fig. 18: TINTE simulation results.

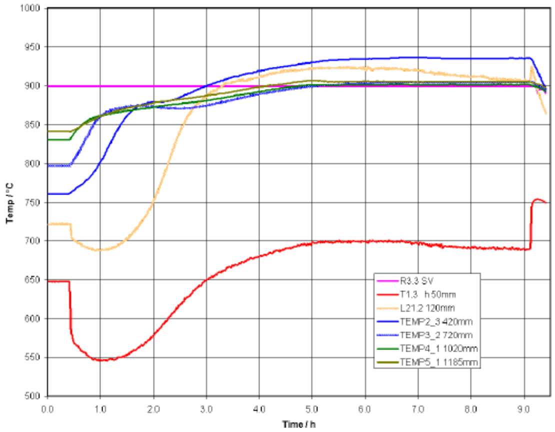


Fig. 19: Overall gas temperatures of the test results.

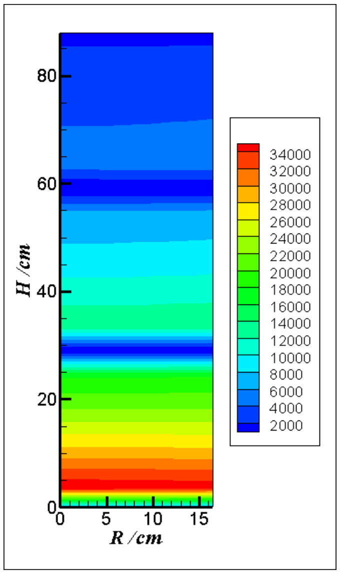


Fig. 20: Graphite corrosion distribution at 9 hr [mol/m^3].

Table 3 lists the weight change of the graphite block derived in the test. The total weights at installation and extraction are 129.538 kg and 118.794 kg respectively. It can be seen that most of the corrosion takes place at the bottom block. The weight differences at bottom, middle and top are 18.79%, 4.14% and 1.95% respectively, while the average difference is 8.29%.

In TINTE simulation, the total graphite mass of these three layers at 0 hr is 120.86 kg and the weight loss is 12.02 kg at 9 hr. The average difference is 9.95 %. While in THERMIX/REACT simulation, the weight loss at 9 hr is about 1100 mol, namely 13.2 kg, and the average difference is 10.92%. The calculation results accord well with the experiment.

Fig. 20 shows the graphite corrosion distribution of TINTE simulation at 9 hr.

Nr.	Type	Layer	Weight at installation [kg]	Weight at extraction [kg]	Difference [%]
1	SGL	bottom	10.470	7.930	24.3
2	SGL	bottom	11.418	10.110	11.5
3	UCAR	bottom	10.240	7.800	23.8
4	UCAR	bottom	11.060	9.232	16.5
5	SGL	middle	10.568	10.198	3.5
6	SGL	middle	11.414	10.900	4.5
7	UCAR	middle	10.240	9.888	3.4
8	UCAR	middle	11.044	10.490	5.0
9	SGL	top	10.492	10.408	0.8
10	SGL	top	11.426	11.134	2.6
11	UCAR	top	10.148	9.996	1.5
12	UCAR	top	11.018	10.708	2.8

Table 3: Graphite weight before and after the test.

V. CONCLUSION

The simulation results show that SPECTRA, TINTE and THERMIX codes are capable to model the NACOK experiment with a good agreement, and the preliminary code-to-code validation is also successfully carried out. The analysis results will be further investigated for study of the fundamental phenomena and prediction of the consequences of such an air-ingress accident. Besides, more research work, including parameter sensitivity analysis, modeling refinement, et al, should be performed to improve the simulation accuracy in the future.

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