



EUROPEAN
COMMISSION

Community Research

RAPHAEL

Contract Number: **516508 (FI6O)**



DELIVERABLE (D-ST3.1)

OXIDATION OF GRAPHITE BLOCKS BY AIR CIRCULATING BY NATURAL CONVECTION.
ELEMENTS FOR THE MODELLING OF AN AIR INGRESS ACCIDENT IN A VHTR.
DEFINITION OF THE NACOK EXPERIMENT.

Author:

L. BRISSONNEAU (CEA)

Reporting period: 15/04/2007– 14/04/2008

Date of issue of this report : 17/12/2007

Start date of project : 15/04/2005

Duration : **48 Months**

Project co-funded by the European Commission under the Euratom Research and Training Programme on Nuclear Energy within the Sixth Framework Programme (2002-2006)

Dissemination Level

PU	Public	
RE	Restricted to the partners of the RAPHAEL project	X
CO	Confidential, only for specific distribution list defined on this document	

RAPHAEL (ReActor for Process heat, Hydrogen And ELectricity generation)



ReActor for Process heat, Hydrogen And Electricity generation (RAPHAEL)		
Sub-project: SP-ST Work package: 3	RAPHAEL document no: RAPHAEL-0804-D-ST3.1	Document type: D D=Deliverable
Issued by: CEA Internal no.: DTN/STPA/LPC/2007/65		Document status: FINAL

Document title						
OXIDATION OF GRAPHITE BLOCKS BY AIR CIRCULATING BY NATURAL CONVECTION. ELEMENTS FOR THE MODELLING OF AN AIR INGRESS ACCIDENT IN A VHTR. DEFINITION OF THE NACOK EXPERIMENT						
Abstract						
In the framework of the RAPHAEL Project, the Sub-Project SAFETY is concerned by the safety problems arising in a block type HTR reactor. In particular, in the workpackage WP3, the behaviour of the core material in case of large air ingress is questioned. In the case of a large leak in an HTR reactor, caused for example by a double break of the co-axial duct between the core vessel and the gas turbine (or steam generator), a large volume of air can enter in the core. As most of the core components are made of graphite blocks, it has to be checked that the oxidation of the blocks would not lead to high levels of radio-contaminant release. In case of double break, the air circulation through the reactor is due to the chimney effect, which is very difficult to model. The NACOK facility set in the Institute for Safety Research and reactor Technology at FZJ can help to determine the oxidation behaviour of graphite blocks with air circulation driven by chimney draught. A NACOK experiment is proposed to yield data for the validation of the REACT/THERMIX code in order to be able to model the oxidation of VHTR graphite blocks in case of air ingress, when the circulation of air is driven by chimney draught. The conditions are set in such way that the graphite temperature is about 850°C and the air flow rate lower than 5 gs⁻¹. The description of the graphite blocks is given; the instrumentation of the experiment and the post test examinations are detailed. This document was checked by H. NIESSEN (FZJ) concerning the technical part						
Revisions						
Rev.	Date	Short description	Author	WP Leader	SP Leader	Coordinator
			L. Brissonneau CEA	H.F. Niessen FZJ	C. Angulo SUEZ-Tractebel	E. Bogusch AREVA NP GmbH
00	10/04/2008	First issue				



**Oxidation of graphite blocks by air
circulating by natural convection.
Elements for the modelling of an air
ingress accident in a VHTR.
Definition of the NACOK experiment.**

L. BRISSONNEAU

DEN/DTN/STPA/LPC/2007/65

***Ce document est le produit d'activités menées dans le cadre d'un système de
management de la qualité certifié ISO 9001:2000 par l'AFAQ***

Date 26/11/2007

Nombre de pages : 2/21



Identification	Technical Note DTN/STPA/LPC/2007/65
Titre	Oxidation of graphite blocks by air circulating by natural convection. Elements for the modelling of an air ingress accident in a VHTR. Definition of the NACOK experiment.
Auteur(s)	L. Brissonneau

Résumé :

In the framework of the Raphael Project (6th FPEC), the “safety” sub-project is concerned by the safety problems arising in a block type HTR reactor. In particular, in the WP3, the behaviour of the core material in case of a large air ingress is questioned. In the case of a large leak in an HTR reactor, caused for example by a double break of the co-axial duct between the core vessel and the gas turbine (or steam generator), a large volume of air can enter in the core. As more of the core components are made of graphite blocks, it has to be checked that the oxidation of the blocks would not lead to high levels of radio-contaminant release.

In case of double breach, the air circulation through the reactor is due to the chimney effect, which is very difficult to model. The NACOK facility, set in the Institute for Safety Research and reactor Technology, at FZJ, can help to determine the oxidation behaviour of graphite blocks with air circulation driven by chimney draught.

A NACOK experiment is proposed to yield data for the validation of the REACT/THERMIX code in order to be able to model the oxidation of VHTR graphite blocks in case of air ingress, when the circulation of air is driven by chimney draught. It is proposed that the conditions are set in such way that the graphite temperature is about 850°C and the air flow rate lower than 5 g.s⁻¹. The description of the graphite blocks is given, as well as the instrumentation of the experiment and the post test examinations are detailed.

This document was checked by H. NIESSEN (FZJ) concerning the technical part.

Mots clés	Safety, HTR , graphite, Oxidation, Natural Convection, NACOK, Raphael
------------------	---

DOB : DDIN	Projet : RCGMA
Domaine : SF	Partenaire(s) : UE

La diffusion des informations contenues dans ce document auprès d'un organisme tiers extérieur au CEA est soumise à l'accord du Chef de Département.

	Nom	Visa
Rédacteur(s)	BRISSONNEAU	
Vérificateur (chef de laboratoire)	POLETIKO	
Emetteur (chef de service)	MAGUIN	

Classification				
DR	CC	CD	SD	Sans
				X

Commissariat à l'énergie atomique
Centre de Cadarache - CEA/DEN/CAD/DTN/STPA/LPC– Bât. 208 - 13108 Saint-Paul-lez-Durance
Tél : 33 - 04.42.25.42.58 - Fax : 33 - 04.42.25.72.87 – E-mail : laurent.brissonneau@cea.fr

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	3/23
	Identification	Indice	Page

LISTE DE DIFFUSION :

Complete document :

Tractebel Engineering, Nuclear Department, Brussels

J. PIRSON

Forschungszentrum Jülich

H. NIESSEN

Areva NP GmbH, Erlangen

G. BRINKMANN

DEN/GRE/DER/SSTH

G. GEFFRAYE

DEN/SAC/DDIN

P. YVON

JC ROBIN

DEN/CAD/DTN/Dir (1 exemplaire)

DEN/CAD/DTN/STPA

J-C MAGUIN

DEN/CAD/DTN/ STPA/LPC

L. BRISSONNEAU

Ch. POLETIKO

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	4/23
	Identification	Indice	Page

Révisions

Date	Objet de la révision	Indice
November 2007	Emission initiale	0

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	5/23
	Identification	Indice	Page

Table of content

1. CONTEXT.	6
2. NEEDS.	6
3. REMARKS ON THE EFFECTS OF TEMPERATURE ON THE GRAPHITE OXIDATION.	7
4. PROPOSITIONS FOR THE NACOK EXPERIMENTAL CONDITIONS.	8
4.1. The NACOK facility.	8
4.2. The NACOK instrumentation.	12
5. EXPERIMENTAL CONDITIONS.	16
5.1. Graphite blocks.	16
5.2. Physical parameters.	17
5.3. Test procedure.	18
6. PRE TEST CALCULATIONS.	20
7. POST TEST MEASUREMENTS	20
8. CONCLUSION	23
9. REFERENCES	23

List of figures

Figure 1 : Photography of the NACOK facility in the ISR at FZJ (courtesy FZJ).	9
Figure 2 : Cross view of the NACOK facility with main dimensions (courtesy FZJ).	10
Figure 3 : Schematic drawing of the NACOK test with graphite blocks.	11
Figure 4 : a) general scheme of the facility with the graphite blocks ; b) location of the thermocouples (red) detailed in graphite blocks and c) location of thermocouple (red) and analysis gas tube (green) in the NACOK experiment for VHTR graphite blocks (both types detailed on both sides). (courtesy FZJ).	15
Figure 5 : Top view of graphite blocks after machining A and B hold for the different graphite grades and 1 and 2 for typical block reactor channels and bigger size channels respectively (provided by FZJ).	17
Figure 6 : Schematic drawing of sampling of the graphite blocks for metallographic examination (scale 1/3).	22

List of tables

Table 1 : comparison of a VHTR block reactor and NACOK.	18
---	----

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	6/23
	Identification	Indice	Page

1. CONTEXT.

In the case of a large leak in an HTR reactor, caused for example by a double break of the co-axial duct between the core vessel and the gas turbine (or steam generator), a large volume of air can enter in the core. As more of the core components are made of graphite blocks, it has to be checked that the oxidation of the blocks would not lead to high levels of radio-contaminant release. In case of double breach, the air circulation through the reactor is due to the chimney effect, which is very difficult to model. The NACOK facility, set in the Institute for Safety Research and reactor Technology, at FZJ, can help to determine the oxidation behaviour of graphite blocks with air circulation driven by chimney draught. It should be underlined that such considerations stands also for the AGR (Advanced Gas Reactor).

In case of a double break of the co-axial duct, after scram, depressurization (of helium) will occur and a mixture of helium and air will enter at the bottom of the core. At the beginning of the accident, the temperatures are : about 1000°C at the bottom of the core and about 400°C at the top of the core. Due to the residual heat from the fuel, the temperatures will increase slowly (because of the large mass of graphite in the core) and tend to homogenize also slowly (because of the low thermal conductivity of the irradiated graphite). As the temperature increases, the irradiation defects of the graphite are annealed and the thermal conductivity increases, leading to a better homogenization and heat removal. Also the residual heat will decrease rapidly.

As helium is lighter than air, the replacement of helium by air in the core, leading to the natural cooling might be quite long (“diving bell” effect of several hours, according to previous NACOK experiments [1]). When the air will circulate, the graphite will be cooled down but as the oxidation reaction by O₂ is exothermic, the temperature could also locally increase. In some hot parts of the core, the CO₂ formed by the oxidation reaction could also oxidized the graphite in CO and –as this reaction is endothermic- lead to cool them.

As the time when chimney draught is established is very sensible to small changes in parameters, it should be consider from a conservative point of view that there is no “grace delay” and thus that air will circulate in the reactor at the very beginning of the accident.

2. NEEDS.

It should always be kept in mind that oxidation of the core will remain limited as 1 m³ of air (at atmospheric pressure) can only lead to the maximum gasification of 0.2 kg of graphite [2], comparing with the total mass of the fuel blocks (more than 100 metric tons). Nevertheless, when air entry would occur, the 4 following points should be checked :

- 1) The air contributes to the cooling of the reactor (versus e.g. exothermic oxidation reaction).
- 2) The corrosion will not damage the support function of the core. It has been shown that the Young’s modulus and the compression strength will decrease by 40% and bend strength will decrease by 55% *when only 8% of graphite is gasified* [3].
- 3) The air will not be in contact with fuel (leading to the oxidation of the pyrocarbon of the fuel and then the increase of fuel temperature).
- 4) The burning limit of CO (depending on gas composition) will not be reached.

A fifth point could also be the production of particles due to the oxidation, as they can be vehicles for the fission product transport [2].

Thus, the local temperature increase (gas and solid phases), the mass of gasified carbon and the composition of the gas phase, when oxidation occurs during the air draught, must be known. This is the purpose of the REACT/THERMIX code to yield such data. The NACOK experiment we propose

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	7/23
	Identification	Indice	Page

in the framework of the Raphael Project has to provide data in order to validate the availability of the REACT/THERMIX code to model the oxidation behaviour, and then can give answers to the four above questions, in the case of an air ingress driven by chimney draught.

As at least two grades of nuclear graphite from two different providers (PCEA from UCAR and NBG17 from SGL) are considered as potential material for the blocks, it is also necessary to check that if clear differences in oxidation behaviour between them can be shown. In effect, oxidation kinetics of the graphites are known from thermo-gravimetric experiments in forced flow and isothermal conditions on small samples. In big blocks, the differences in oxidation kinetics between graphite can lead to differences in mass loss but also to differences in microstructural changes (in particular the porosity network on which depends the "tightness" of the graphite). The differences in mass loss must be found by the modelling. But it has to be verified that the microstructural changes are also comparable and if not, it has to be stated what grade of graphite is most sensible to the oxidation (at a given burn-off).

Last but not least, if the experiment is generic for code validation, it should not be too far from ANTARES conditions (flow rates, pressure drops...).

3. REMARKS ON THE EFFECTS OF TEMPERATURE ON THE GRAPHITE OXIDATION.

It is probable that the temperature of the reactor core will be higher than 800°C, and in some cases it could attain values as high as 1600°C.

It has been underlined by many authors that the support function and the tightness of the graphite (points 2 and 3 –and thus also 5- of the former chapter) are particularly sensitive to oxidation *around 800°C, when oxidation is fast but still takes place in the porosity of the graphite*. This is particularly true if the binder of the graphite is corroded faster than the filler (coke). Depending on the grain size of the graphite (and then the quantity of binder) the oxidation can cause more or less damage : graphite with bigger grains will be more sensitive to binder oxidation as it would lead more easily to grain removal.

All the parameters play an important role in the oxidation rate of the binder : the temperature, the quantity of oxygen, the pressure, the flow rate, the burnup and the nature of the binder. The results of oxidation rates obtained from TGA experiments with forced flows on small samples can not be straightforwardly applied for the helium channels. Moreover, the literature shows that great discrepancies can be found for different graphite grades at these temperatures ([4] or more recently [5] on graphites of interest).

Thus a specific study of the two graphite grades in the NACOK facility around 800°C seems important. The post-test metallographic examinations, as well as specific surface and porosity measurements on the blocks are also of interest (see § 7).

Above 1100°C, the oxidation by air will be very fast and occur only on the surface : O₂ reacts before it diffuses in the porosity. Oxidation by CO₂ could occur in turn, generating corrosion in upper parts of the reactor and contributing to cooling due to the endothermic nature of this reaction. It will also increase the CO content at the exit (here point 4 of the former chapter is concerned).

At 1600°C, it is probable that air will react in the very first cms of the blocks (the height depending on the air flow and the velocity), yielding mostly CO. In that case, metallographic examinations will yield few information (except for a upper CO₂ oxidation ?). The most interesting information would be the real flow rate in the channels and the height of gasified graphite (the total mass is given by weight but the location of the reaction is not known and might also depends on the graphite grades, although to a lesser degree).

Few data from TGA experiments are relevant at these high temperatures as mass transfer is the limiting process. In both cases (low and high temperatures) the gas analysis at different heights

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	8/23
	Identification	Indice	Page

would help to understand how the reaction evolves along the channels. In both cases also, the volume and the location of the gasified graphite will be strongly related to the thermo-hydraulic conditions (mainly mass flow and velocity).

Though experiments at the highest temperature could be of interest, it is probable that the most probable and damaging case concerns the “low temperature” oxidation.

It is our feeling that most valuable information will be gained by an experiment where most of the graphite is at a temperature of, or lower than, 850°C. Moreover, the upgrade of the facility in order to perform experiments at temperatures as high as 1100°C would lead to unbearable costs and delays.

4. PROPOSITIONS FOR THE NACOK EXPERIMENTAL CONDITIONS.

4.1. The NACOK facility.

The NACOK (Naturzug in Core mit Korrosion) facility is a 8 m high, 300 mm side, chimney made of steel tubes, heated by outer heating elements (see Figure 1 for a photography of the facility and Figure 2 for its general cross-section view). The blocks of graphite will be placed on ceramic supports at the base of the column. Three 300 mm high blocks of each type (4 types used) are set one over the other from the top of the facility. Blocks are instrumented with systems for temperature measures and gas phase analysis (see § 4.2). Above blocks, ceramics pellets are set to a height such that the induced pressure drop will allow to reach the approximately desired mass flow. The pellets laid on a steel plate with 8 mm diameter holes for the air circulation. The maximum height for the pellets is 300 mm. A schematic drawing of the facility with the graphite blocks test is given in Figure 3 (see also Figure 4 for more detailed informations).

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	9/23
	Identification	Indice	Page



Figure 1 : Photography of the NACOK facility in the ISR at FZJ (courtesy FZJ).

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	10/23
	Identification	Indice	Page

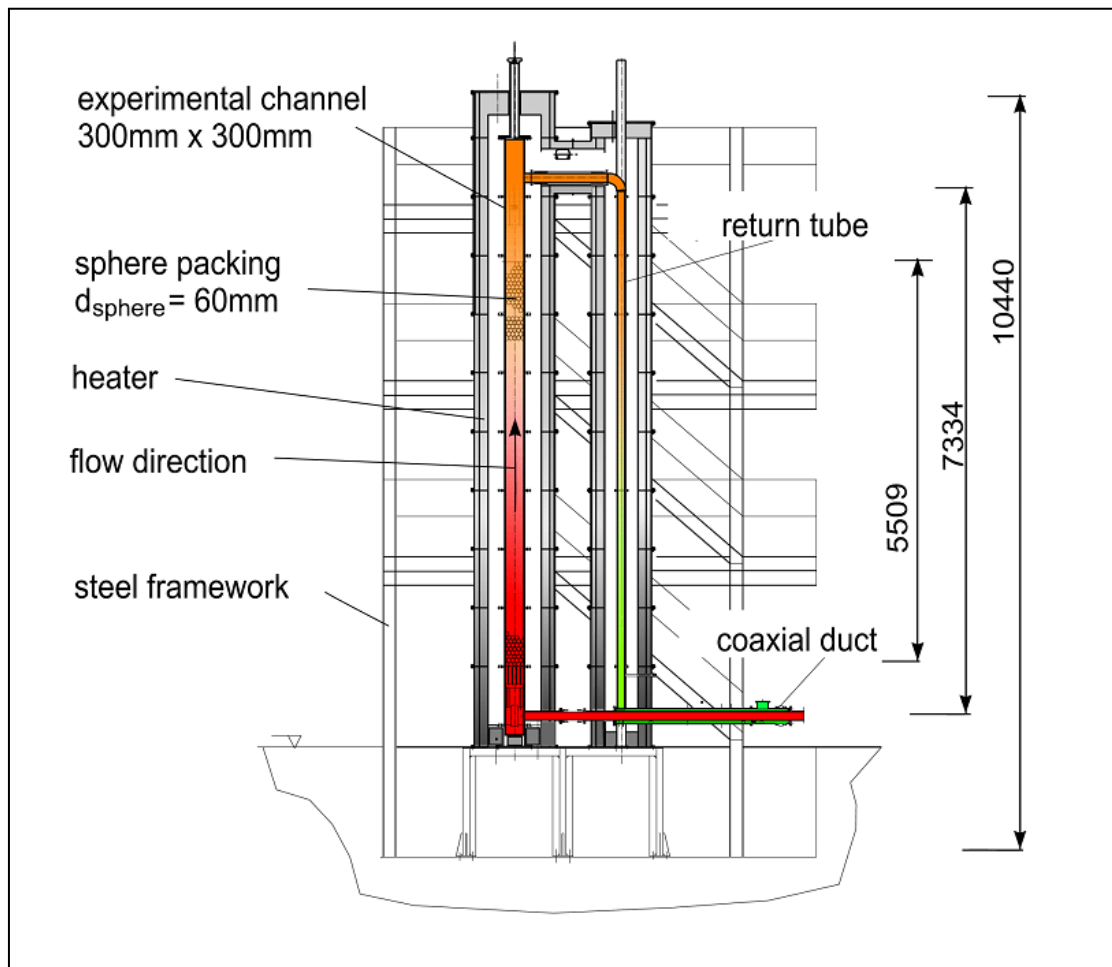


Figure 2 : Cross view of the NACOK facility with main dimensions (courtesy FZJ).

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	12/23
	Identification	Indice	Page

4.2. The NACOK instrumentation.

In NACOK, many probes can be inserted in or between the blocks or in the column in order to measure :

- the temperature ;
- the gas composition ;
- the gas flow rate ;
- the pressure.

The temperature is measured by thermocouples Ni/NiCr on the outer surface of the column and in the graphite blocks. The measure of the temperature allows to determine the cooling by air in natural convection and the increase in temperature in the graphite due to the exothermic or endothermic reactions.

The temperature will be measured :

- At the entry of facility (cold part)
- at different heights in the blocks with a thermocouple able to move up and down in a hole in the graphite or between the blocks.
- At the entry and the exit of each block
- at the exit of the chimney.

The total number of thermocouples in the graphite blocks will be 23. The measures are recorded every minute.

The gas flow rate is measured at the inlet by a rotating piston flow meter.
The accumulated flow is recorded every minute.

The gas analyses is performed by continuous sampling of the gas phase in 5 mm diameter tubes that lead to the following analysers :

- Infrared analyser (URAS) for CO and CO₂.
- Thermal conductivity analyser (CALDOS) for CO and CO₂.
- Magnetic analyser (MAGNOS) for O₂.

It is necessary to limit the gas phase analyses in order to no to disturb the natural flow.

5 gas sampling tube will be placed :

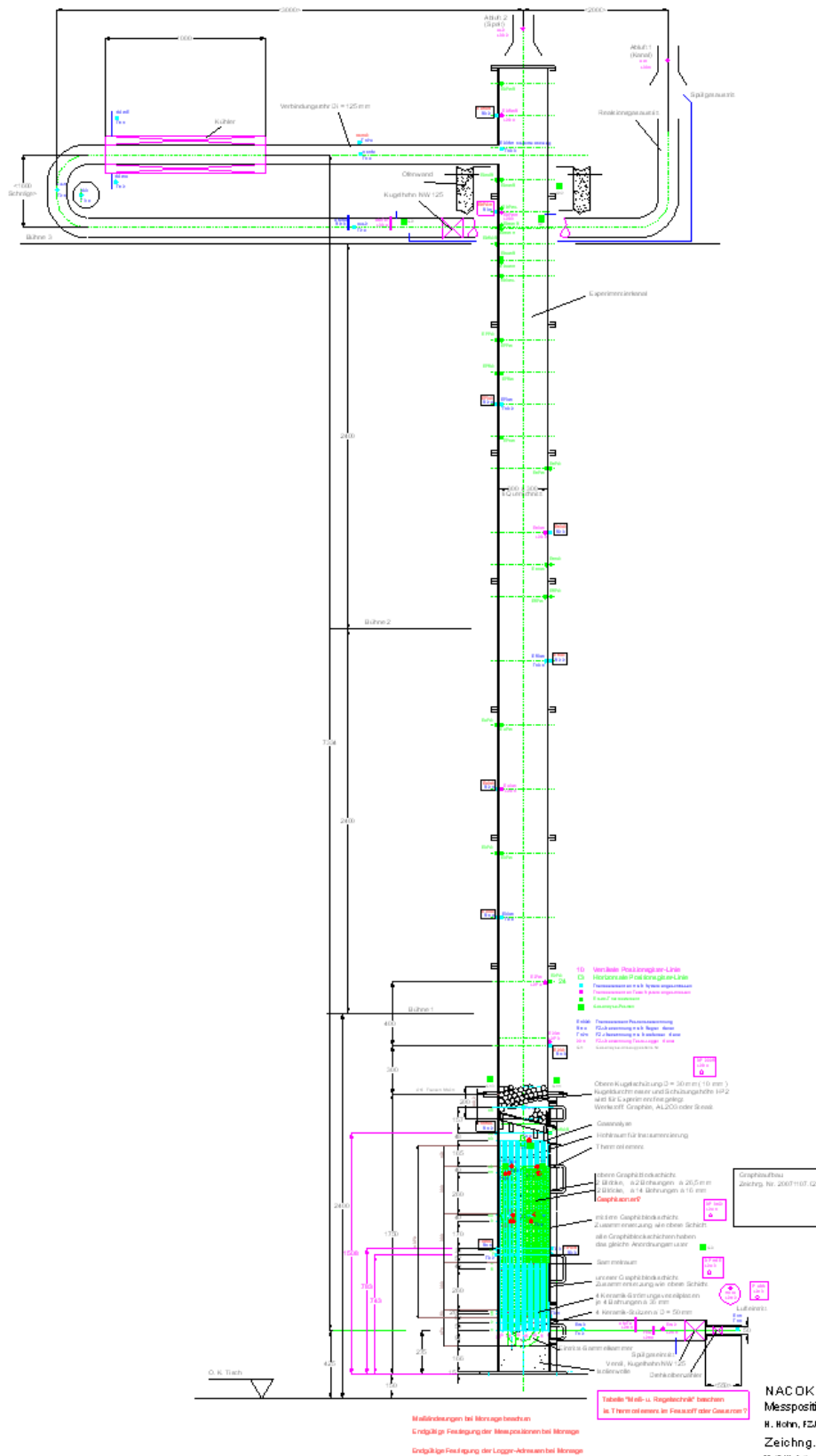
- Two at the top of the bottom blocks.
- Two at the top of the medium blocks.
- One at the top of the top blocks.

The measures are recorded every 5 minutes.

The pressure is determined from some of the analysis tubes. Pressure values will be given every hours, in additional documents.

The data are delivered in the form of Excel sheets. Data are treated by FZJ so that only the interesting values are available (e.g. actual temperature at experiment time ...). The planned position of the measuring points may differ from the real positions because of practical reasons. These should be considered when comparing between pre and post test simulations.

The location of the instruments is given in Figure 4, as well as a general description of the experiment to be conducted.



b)

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	15/23
	Identification	Indice	Page

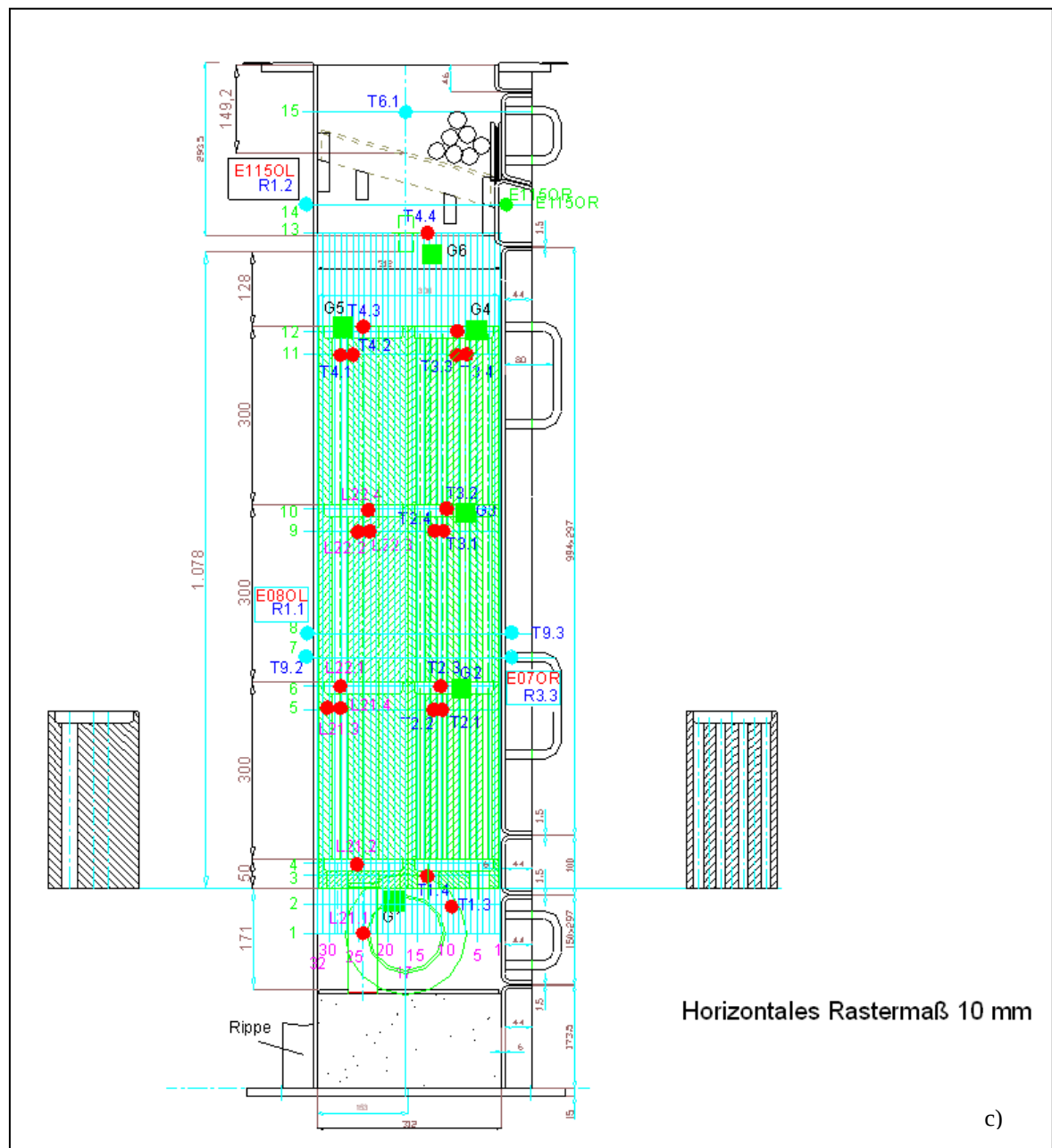


Figure 4 : a) general scheme of the facility with the graphite blocks ; b) location of the thermocouples (red) detailed in graphite blocks and c) location of thermocouple (red) and analysis gas tube (green) in the NACOK experiment for VHTR graphite blocks (both types detailed on both sides). (courtesy FZJ).

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	16/23
	Identification	Indice	Page

5. EXPERIMENTAL CONDITIONS.

5.1. Graphite blocks.

Four types of graphite blocks will be used in the experiments, made of two graphite grades with two channel diameters.

The graphite grades are those studied in the material program RAPHAEL M. They are from two different providers, SGL and GrafTech UCAR. These are :

- NBG-17 grade from SGL,
- PCEA grade from GrafTech UCAR.

The blocks used in the experiment must have been chemically purified so that they correspond to “nuclear graphite” grades, with ash content lower than a given value, typically 200 ppm. As the ash content can greatly influence the oxidation kinetic, especially at lower temperatures, the purity of the graphite must be close to the one studied in the Material program, or by the CEA, so that the kinetic data obtained from TGA experiments can be input in the REACT code. Also, the physical data of the grades must be known to be input in the THERMIX code. Thus the final choice of the grades will depend on the availability of the materials and of their physical data (for example NBG 18 could be chosen instead of NBG 17).

The graphite blocks (Figure 5) are square base 15x15 cm², 30 cm high. Considering a density of about 1.8, the mass of one block is about 12.1 kg.

14 or 2 channels of diameter 16 or 26.5 mm respectively will be machined, so that the pressure drops being the same in both geometries. The 16 mm channels are typical of the helium channels in the ANTARES design. After machining, the mass of one block is about 10.5kg.

In both geometries the pressure drop are identical, so the mass flow of air should be the same. But the surface of graphite in direct contact with air will be different :

- 2111 cm² in the small holes blocks, 150 cm² per hole ;
- 500 cm² in the big holes blocks, 250 cm² per hole.

Thus the mass flow of air in the big holes per unit surface of graphite is approximately 4.2 times the flow rate in the small holes. It will be an important point to check if the oxidation rate varies according to this factor. The decrease of oxygen content in the small holes blocks could be faster and then the oxidation gradient sharper.

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	17/23
	Identification	Indice	Page

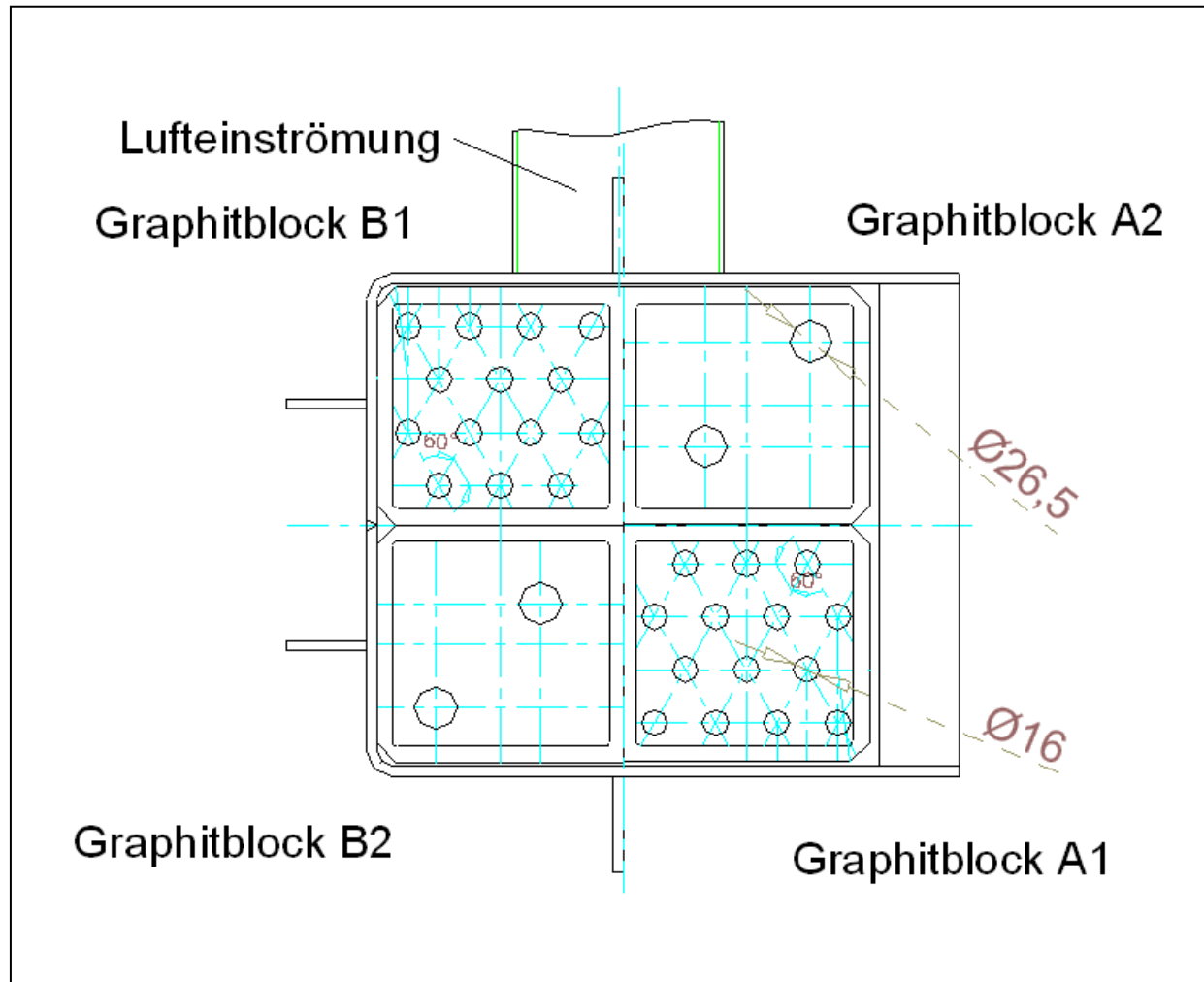


Figure 5 : Top view of graphite blocks after machining A and B hold for the different graphite grades and 1 and 2 for typical block reactor channels and bigger size channels respectively (provided by FZJ).

5.2. Physical parameters.

The most important parameters to be set in the experiment are the temperature and the flow rate. In order to get a temperature of 850°C in most part of the graphite, an external temperature of 900°C must be applied on the steel column. From FZJ experience, when the temperature is set to 900°C on the outer side of the column, the average temperature of the graphite blocks should be 850°C, and about 700°C at the base of the bottom blocks, because of the heat loss through the ceramic support.

The flow rate can not be accurately controlled, but from former NACOK experiments, it can be estimated that a flow rate of air of 5 g.s^{-1} can be achieved by a ceramic pebbles height of 30 cm and a flow rate of 15 g.s^{-1} when no ceramic pebbles are placed in the column.

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	18/23
	Identification	Indice	Page

In order to be as close as possible of the ANTARES parameters, it was chosen to set the flow rate to be as low as possible, ie 5 g.s^{-1} .

In the following table (from [6]), a VHTR block reactor and NACOK configurations are compared.

		Block reaction:	NACOK	NACOK
Core Inner Diameter	Dci / [m]	2.84		
Core outer diameter	Dca / [m]	4.84	0.3 (square)	0.3 (square)
Core height	Hc / [m]	8	8	8
Core sectional area	Ac / [m ²]	12	0.09	0.09
Core outlet temperature	THG / [°C]	850	900	900
Core inlet temperature	TKG / [°C]	450	20	20
Buoyant height	Ha / [m]	12	8	8
Diameter of cooling channels	dgb / [m]	0.016	0.016	0.0265
Pitch of cooling channels	tbg / [m]	0.038	<0.038	<0.038
Average temperature for accident conditions	TmA / [°C]	800 ?	≈800	≈800
estimated air mass flow at DBA	m / [g / s]	50	5	5

Table 1 : comparison of a VHTR block reactor and NACOK.

The duration of the experiment depends on the rate of consumption of the graphite blocks, which is estimated from the gas phase analysis. The experiment will stop after approximately 10% of the graphite is consumed. 5 g.s^{-1} of air can gasified at most $0,75 \text{ g.s}^{-1}$ of graphite (with CO_2 as reaction product, twice more by producing CO), corresponding to 27 kg of graphite in 10 hours. The total mass of the 12 graphite blocks should be around 125 kg. The experiment duration must be longer than 4 hours and should not exceed 20 hours (if less than 20% of oxygen react).

5.3. Test procedure.

The general progress of the experiment is the following. After placing the graphite blocks and installing the instrumentation, the column is filled with N_2 . Instrumentation is checked, and before the beginning of the heating, the recording of the parameters, temperature, gas phase composition,

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	19/23
	Identification	Indice	Page

is started. The column is heated by outer heating elements (see Figure 2) until a temperature of 900°C is obtained on the outer surface of the column and that a stable temperature profile is obtained in the graphite. Then inlet tube is opened, the exhaust valve located at the top of the column is opened and air is allowed to enter at the bottom of the column. Constant evaluation of mass loss is performed from the gas phase analysis. The experiment will be stopped when about 10% of total mass of graphite is gasified.

The complete procedure for the experiment could be the as follows [6] :

1. Perform a leak test using nitrogen i.e. pressurization test with time dependent pressure decay.
2. Increase temperature in the main test section from ambient to 300 °C at a rate of 60°C/h.
3. Check the temperature indications during this process for possible inconsistencies and make corrections if necessary (hardware and software).
4. Perform a leak test.
5. Introduce ambient air for 3h. (First test)
6. Test the mass flow meter during this process and check for possible graphite corrosion.
7. Increase temperature in the main section from 300°C to 350°C at a rate of 60°C/h.
8. Repeat 5. and 6. (Second test)
9. Increase temperature in the main section from 350°C to 400°C at a rate of 60°C/h.
10. Repeat 5. and 6. (Third test)
11. Increase temperature in the main section from 400°C to 450°C at a rate of 60°C/h.
12. Repeat 5. and 6. (Forth test)
13. Increase temperature in the main section from 450°C to 500°C at a rate of 60°C/h.
14. Repeat 5. and 6. (Fifth test)
15. Decrease temperature to 400°C (weekend).
16. Increase the temperature in the main test section to 900°C at a rate of 60°C/h.
17. Check all instruments and make corrections if and where necessary.
18. Allow approximately 8 hours for the system to stabilize.
19. Open the system to atmosphere. (Sixth test, MAIN TEST)
20. Measure stabilizing natural draught flow rate.
21. Close air inlet after a approximate corrosion of 10% to 11% has been reached.
22. Fill the system with nitrogen.

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	20/23
	Identification	Indice	Page

23. Reduce the temperature in the complete test section to 300°C.
24. Carry out a leak test.
25. Reduce the temperature in the complete system to ambient temperature.
26. Carry out a leak test at room temperature.

6. PRE TEST CALCULATIONS.

Pre test calculations by REACT/THERMIX have to be performed before the start of the experiments. The results will be useful to forecast the duration of the experiment and to confirm that the parameters chosen for the NACOK experiment will permit to yield the necessary data for the validation of the code.

The kinetics data of the graphite obtained from thermo-gravimetric experiments will be used. As the code is 2D, at least 2 calculations must be performed in order to take into account the difference between the two types of graphite or of the geometry of the blocks.

The critical review of the data and the calculation by REACT/THERMIX are to be given in an other report [7].

7. POST TEST MEASUREMENTS

The following post test examinations will be conducted :

- mass loss determination ;
- measures of channels dimensions ;
- porosity (helium pycnometry) ;
- specific surface (BET) ;
- metallographic examination (optical microscope, SEM) ;
- additional tests are done by the graphite producers (in the framework of the ML sub-project).

The REACT/THERMIX code will yield the temperature of the solid and the gas phases, the composition of the gas phase and the mass of gasified graphite at each step time of the calculation at the different nodes.

These calculations have to be compared with the data obtained in the NACOK experiments. The temperature and the composition of the gas phase will be recorded continuously but in a limited amount of places.

Concerning the mass of gasified graphite, an estimation is done continuously from the analysis of the gas phase. But the corrosion rates obtained will be checked by a balance between the calculated mass from gas phase analysis and the final mass of the blocks (adding "graphite dust" collected at the bottom of the column if necessary).

The mass of each block must be carefully weighed after test and compared to its initial mass. The total mass of collected graphite dust must be also weighed. These are *the most important measures* to be done after test.

Then, the following examinations might yield interesting additional information on the oxidation process and its consequences. The metallographic examinations will provide information on what part of the oxidation concerns the external surface of the channels and what part concerns inner graphite. The enlargement of the channels might lead to change in the gas flow behaviour, when inner graphite corrosion is damaging for the mechanical integrity and might provide short-circuit to

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	21/23
	Identification	Indice	Page

the fuel channels (distance between helium channels and fuel channels may be as short as 3 or 4 mm).

The measurement of the channel dimensions (diameter at the bottom and the end of the blocks) will yield information on the mass of gasified graphite at the outer surface of the channels.

After these measurements are done, the blocks will be cut for the remaining examinations. Great care should have to be taken during the cutting and machining of the samples, because it is expected that the graphite would have lost a significant amount of its mechanical strength due to the inner oxidation. In Figure 6, a *proposal* of sampling is given. Some samples will be used for pycnometric volume and BET surface measurements and the other for microscopic examinations. The ideal size of the sample is a cube of 0.7 cm side for direct BET measurements. But bigger samples can be used for helium pycnometry and microscopy. It is proposed that slices with a base of 0.7x0.7 cm² are cut between channels. The definite cut will be decided after examinations of the blocks after the test.

The samples should be machined in such a way that regular shapes are obtained so that apparent density can be easily obtained from easy dimension measurements and weights.

The pycnometric density measurements allows to directly calculate the open porosity of the graphite. The close porosity is deduced from comparison with the theoretical density of graphite (apparent density being also known). The oxidation in the pores can lead to the opening of the porosity which means a more direct route towards fuel. The opening of the porosity is estimated by comparing two samples of same dimensions before and after oxidation. It will be of interest to compare the volume of open porosity as a function of the depth from a channel. It is necessary for this purpose to dispose of some few graphite samples from the blocks before their oxidation (machining drops).

The BET measurements yields the specific surface of the sample. It is known that the specific surface greatly increases during the oxidation, partly due to the opening of the porosity. Depending on the regime of oxidation (on the surface or in the pore) the surface increase is more or less pronounced. Though of less interest than pycnometric density, some few measures should be performed, which should yield some interesting information on the local oxidation conditions.

SEM and optical microscope examination can help to determine what it the depth of most affected graphite, as well as the microstructure of the remaining graphite. The preferential attacks between coke grains can be put into evidence by this mean.

Samples are first examined in direct view by SEM and then cross sectional examinations by optical microscopy and SEM will be performed. In order to realize the cross section, the samples are molded in resins, cut and polished.

Considering the large amount of samples proposed from Figure 6 (six kinds of sampling per graphite blocks, ie 36 trenches for microscopy and as much for BET and pycnometry), it is proposed that, at first, only the samples taken at the top of each blocks should be examined, and also all those of one block. In case of major differences between blocks, the other examination could be performed.

Hence, at first, only 15 pycnometrics experiments and metallographic examinations are planned be realized. This amount may also be reduced from first measurements and examinations.

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	22/23
	Identification	Indice	Page

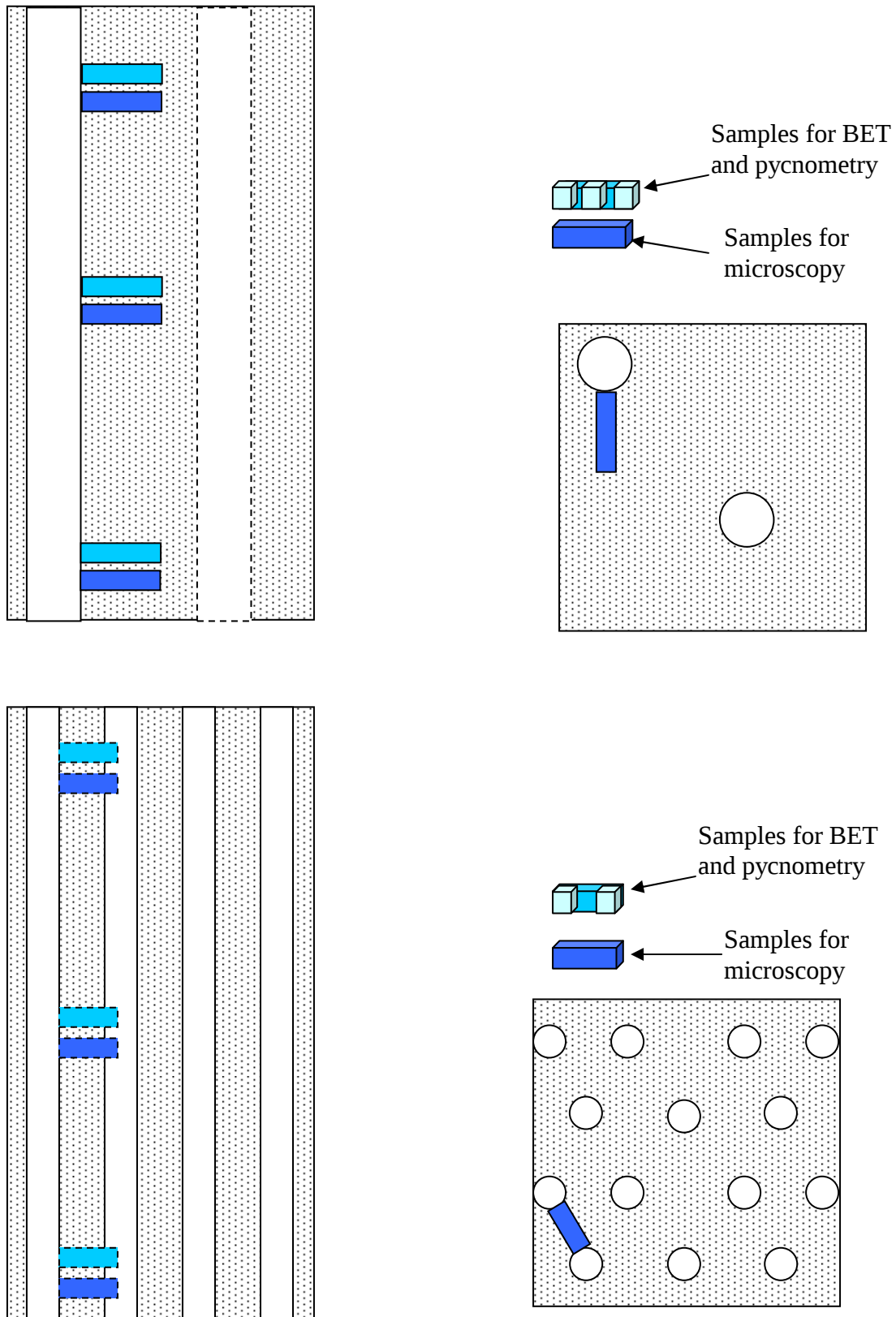


Figure 6 : Schematic drawing of sampling of the graphite blocks for metallographic examination (scale 1/3).

 DEN/DTN/ STPA	NT DTN/ STPA/LPC/2007/65	0	23/23
	Identification	Indice	Page

8. CONCLUSION

A NACOK experiment is proposed to yield data for the validation of the REACT/THERMIX code in order to be able to model the oxidation of VHTR graphite blocks in case of air ingress, when the circulation of air is driven by chimney draught.

It is proposed that the conditions are set in such way that the graphite temperature is about 850°C and the air flow rate lower than 5 g.s⁻¹.

The description of the graphite blocks is given, as well as the instrumentation of the experiment and the post test examinations are detailed.

9. REFERENCES

1. Kuhlmann, M., *Experimente zu Gastransport und Graphitkorrosion bei Luftenbruchstörfällen im Hochtemperaturreaktor*, in *Fakultät für Maschinenwesen*. 2000, RWTH Aachen.
2. Moormann, R. *Phenomenology of graphite burning in massive air ingress accidents*. in *3rd International Topical Meeting on High Temperature Reactor Technology*. 2006. Johannesburg, South Africa.
3. Sato, S., et al., *Degradation of fracture mechanics properties of reactor graphite due to burn-off*. Nuclear Engineering and Design, 1990. **118**: p. 227-241.
4. Lewis, J.B., *Thermal gas Reactions of Graphite*, in *Modern Aspects of Graphite Technology*, L.C.F. Blackman, Editor. 1970, Academic Press, London. p. 129-199.
5. Hinssen, H.-K., et al. *Oxidation experiments and theoretical examinations on graphite materials relevant for the PBMR*. in *3rd International Topical Meeting on High Temperature Reactor Technology*. 2006. Johannesburg, South Africa.
6. Niessen, H.F., *Minutes of the meeting for planning the first NACOK experiment*. 2007, FZJ, Institute for Safety Research and Reactor Technology, Jülich.
7. Haque, H., *REACT/THERMIX pre calculations of the NACOK experiments with ANTARES like graphite blocks*. 2008, AREVA NP GmbH, Erlangen.