EUROPEAN COMMISSION
Community Research

RAPHAEL

Contract Number: 516508 (FI6O)

DELIVERABLE (D-ML4.2)

Development of Design Data and Guidelines for Graphite – Scope and Planning

Author(s): **M W Davies (NNC)**

Reporting period: e.g. 15/04/2005– 14/04/2006

Date of issue of this report : 30/06/2006

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 (place an X in the appropriate cell)		
PU	Public	
RE	Restricted to the partners of the RAPHAEL project	
CO	Confidential, only for specific distribution list defined on this document	X

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

Distribution list

Person and organisation name and/or group	Comments
RAPHAEL SP ML Work Package 4 participants	
RAPHAEL SP ML Work Package 3 Participants	
RAPHAEL SP ML Work Package 2 Participants	

ReActor for Process heat, Hydrogen And ELectricity generation (RAPHAEL)		
Sub-project: SP-ML **Work package:** 3	**RAPHAEL document no:** RAPHAEL-0606-D-ML4-2	**Document type:** D=Deliverable / M=Minute / P=Progress report / S=Support
Issued by: NNC(UK) **Internal no.:** 12820/TR/019		**Document status:** Final

Document title

Development of Design Data and Guidelines for Graphite – Scope and Planning

Abstract

Within the RAPHAEL-IP framework, there is a requirement to develop design data and guidelines for the design of the graphite core of a (V)HTR. This report reviews the current situation with regard to graphite data and guidelines on design methodology for the European R&D activities on the (V)HTR, and outlines the overall scope and planning requirements for their development.

Revisions

Rev.	Date	Short description	Author	WP Leader	SP Leader	Coordinator
00	dd/mm/yyyy	First issue	Name, Organisation	Name, Organisation *Signature*	Name, Organisation *Signature*	Name, Organisation *Signature*
01	30/06/2006	First issue	Mike Davies NNC	Derek Buckthorpe NNC	Derek Buckthorpe NNC	E. Bogusch

DOCUMENT ISSUE RECORD
(engineering documents)

Adding value through knowledge

Document Title :	**Development of Design Data and Guidelines for Graphite – Scope and Planning**
Project Reference:	12820

Purpose of Issue : For use

Security Class : Commercial-in-Confidence

Issue	Description of Amendment	Originator/ Author	Checker	Approver	Date
01 Draft	First draft for comment				
		M W Davies	D E Buckthorpe		
01	Checker comments incorporated	M W Davies	D E Buckthorpe	A Cheyne	
		M W Davies	D E Buckthorpe	A Cheyne	

Total number of pages:	Intro:	1 - 7	Text	8 - 16	Tables		Figures		Appendices	A

Previous issues of this document shall be destroyed or marked **SUPERSEDED**

Distribution: SINTER, HTR-M1 Parteners (via SINTER), NNC: M W Davies, D E Buckthorpe, EDMS

3050aJan01 Controlling procedure - QP11, QP40 *Docprodref*

Table of contents

List of Tables

List of Figures

1 Introduction

In order to carry out the design of the graphite core in a (Very) High Temperature Reactor (V)HTR it is essential to have reliable graphite data and a proven design methodology. This needs to take account of the known lifetime degradation mechanisms. With regard to the graphite(s) selected, it is important to have a full characterisation of the as-manufactured material(s) as well as a complete set of information on how particular properties vary with fast neutron irradiation and irradiation temperature over the required range. The oxidation resistance of graphites at high temperature also needs to be determined due to its relevance during normal operation and for licensing and safety analyses of air and water ingress accidents. The need to minimise the activity of the graphite at the end of life in order to facilitate final disposal of the core following decommission will require appropriate limits on, and close control over, the impurity levels in the as-manufactured material(s).

In the past, the design of graphites cores has generally been carried out by the countries in which the reactors were built, using their own developed methodology and data on their particular graphites. The countries in question were the UK, USA, France, Germany and Russia, and more recently Japan. However, most of the graphites that have been used in previous core designs are no longer manufactured commercially, and currently available grades are limited to a few specific sources of raw materials. Therefore, there is a major programme of work underway, as part of the European VHTR R&D activities, to qualify some potential graphites. In particular, long term irradiation experiments are being undertaken in the High Flux Reactor (HFR) at Petten. Few of the graphites being tested may turn out to be suitable for a modular (V)HTR and there may therefore be a need to develop improved graphites in the future.

In addition, the methodologies developed in each country will have similarities, but there will also be clear differences. It is beneficial to have a unified methodology for core design and this is being addressed through the ASME. A Project Team has been established and is developing a consensus design code for graphite components.

This report reviews the current situation with regard to graphite data and guidelines on design methodology for the European R&D activities on the (V)HTR, and outlines the overall scope and planning requirements for their development.

2 Graphite design data

2.1 Selected Graphite Grades

The graphites for the irradiation experiments were selected at specialist graphite meetings. They cover the two main coke sources (petroleum and pitch) and the three main manufacturing methods (extrusion, iso-moulding and vibro-moulding).

Grade	Manufacturer	Coke	Process
PCEA	GrafTech	Petroleum	Extrusion
PPEA	GrafTech	Pitch	Extrusion
PCIB-SFG	GrafTech	Petroleum	Iso-moulding
BAN	GrafTech	Needle	Extrusion
NBG-10	SGL	Pitch	Extrusion
NBG-20	SGL	Petroleum	Extrusion
NBG-25	SGL	Petroleum	Iso-moulding
NBG-18	SGL	Pitch	Vibro-moulding
NBG-17	SGL	Pitch	Vibro-moulding
IG-110	Toyo Tanso	Petroleum	Iso-moulding
IG-430	Toyo Tanso	Pitch	Iso-Moulding

Table 2-1 Selected graphite grades

2.2 Irradiation experiments

Irradiation experiments are required to provide data up to the peak fluence expected[1] and over the appropriate temperature range. For a (V)HTR the temperature range of interest is considered to be 600°C to 1100°C. It is recommended that the data be obtained at 100°C intervals, but since the fluence at the upper and lower temperatures will be small, the data are to be obtained at 100°C intervals between 650°C and 1050°C inclusive. Two experiments are being undertaken in the HFR at Petten. The first stage of this irradiation programme (INNOGRAPH 1A) has already been carried out within the 5th Framework Programme (FP5, HTR-M1). Around 200 small samples covering eight graphites were involved. The samples were first of all pre-characterised, and then irradiated to a maximum of around 1/3 peak fluence at a temperature of 750°C. The test rig has been removed and dismantled and, at the time of writing of this report, around 50% of the samples have already been subjected to some post-irradiation examination (PIE). The measurements are summarised in Ref 1. The peak fluence to be achieved is ~25 displacements per atom (dpa), or ~200 x 10^{20} $n.cm^{-2}$ Equivalent DIDO Nickel Dose (EDND). Thus in INNOGRAPH 1A the

[1] Peak fluence is that appropriate to a pebble bed core design. The replacable components in a prismatic core design should not exceed 1/3 of this value.

samples have been irradiated up to ~9 dpa, or ~70 x 10^{20} n.cm^{-2} EDND. (It should be noted that this would provide sufficient data at this temperature for a prismatic design.) The irradiation to peak fluence will be completed in INNOGRAPH 1B, which is about to commence. Shortly after INNOGRAPH 1A started it was noticed that two of the capsules were only running at ~670^{o}C. This will provide useful data as it is close to the lower temperature value of interest (650^{o}C).

A similar irradiation at 950^{o}C is to be carried out, initially to 1/3 peak fluence, in the FP6 (Raphael). This is referred to as INNOGRAPH 2A. Pre-characterisation measurements on the prepared samples will be available during 2006. Due to the benefits of the data at the lower temperature in the INNOGRAPH 1A experiment, it was decided to run the same capsules at a temperature ~100^{o}C lower, i.e. at ~850^{o}C. Again, although limited, the data should provide a useful indication of the behaviour at this temperature. The second irradiation stage to peak fluence at this temperature (referred to as INNOGRAPH 2B) will be undertaken between 2007 and 2009 and the full set of PIE measurements on the samples at the end of this irradiation will not be available until ~2010. Figure 2-1 gives a schematic of the data ranges to be expected from the different irradiation experiments within HTR-M1 and Raphael-IP.

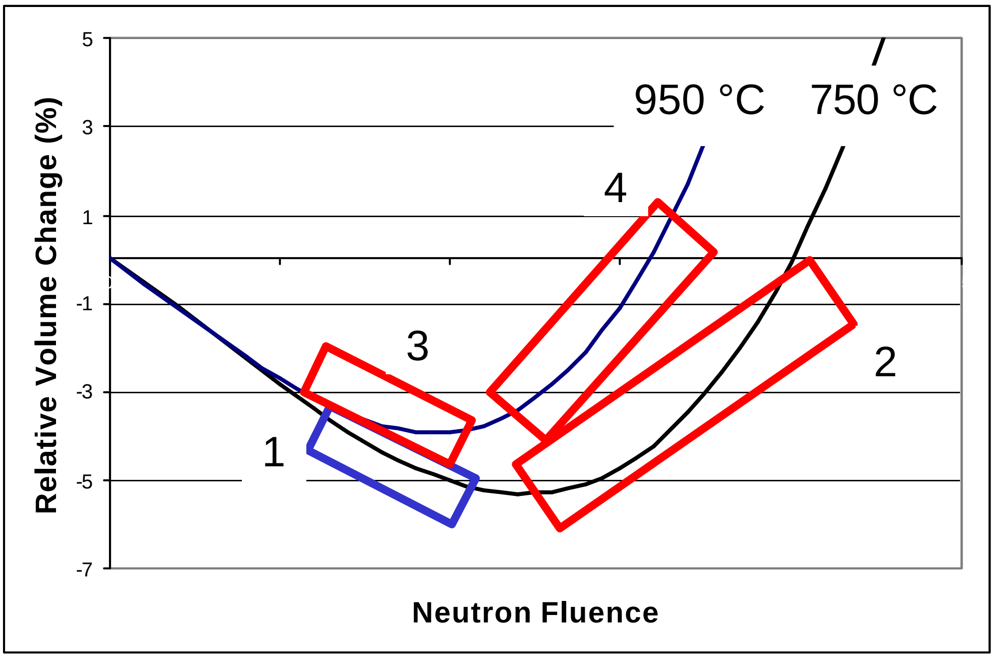

Figure 2-1 Schematic overview of the different irradiation experiments within HTR-M1 and Raphael-IP

The only remaining temperature at which data are required is then 1050^{o}C, and it is understood that a separate experiment is being undertaken in the OSIRIS reactor at this temperature. However, the irradiation is only planned to reach a few dpa.

It will also be necessary to carry out an irradiation creep experiment. Such an experiment is being planned in the USA and it is hoped that this will provide the necessary data, which will be made available through the Gen IV collaboration.

2.3 Graphite irradiation data base

For the pre-characterisation of the samples there was a need to measure their initial dimensions, mass, dynamic Young's modulus (DYM), coefficient of thermal expansion (CTE) and thermal conductivity (via measurements of thermal diffusivity). The sample volumes and densities were calculated from the dimensions and masses. For the irradiated samples subjected to PIE, only their dimensions and DYM have been re-measured to date.

Before the experiment got underway, an Excel worksheet was proposed (comprised of individual spreadsheets for each graphite grade) that would be used to summarise the data in a convenient way and would enable the appropriate 'design curves' to be produce. At the conclusion of the HTR-M1 programme there was a requirement to review the available pre-characterisation and post-irradiation measurements and to enter them into this worksheet. This was carried out in Ref 2. Due to the large amount of data and grades of graphite it was decided to produce a single spreadsheet for each graphite grade. The pre-characterisation data are contained in the first part of the spreadsheet, the irradiated measurements are contained in the second part of the spreadsheet, and the fractional/percentage changes are calculated in the third part.

There was also a requirement to assess the data and to make a comparison with previously irradiated graphites. This was carried out in Ref 3. The calculated changes from the third part of each spreadsheet are used to produce graphical plots of the changes with respect to fluence. This can either be in terms of dpa or EDND. These will be the 'design curves' for the properties in question and polynomial fits to the data can be used for computational models used to calculate through life stresses and distortions in components.

2.4 Tests on oxidation under accident and normal operation conditions:

The limited oxidation resistance of porous C-based materials causes potential problems when they are in contact with oxidising atmospheres. During accident conditions this can lead to a significant localised reduction in strength at exposed surfaces. The necessary safety analyses on graphite oxidation therefore require a detailed knowledge of the oxidation kinetics. Some rapid oxidation tests have been performed on the selected graphites in the FP5 HTR-M/M1 programs [Ref 4 and 5] and further investigations are proposed. These are necessary to determine the behaviour of the different graphites under accident conditions.

Oxidation of the graphite also takes place during normal operation due to impurities in the cooling gases. In this case however the oxidation rate of the graphite is extremely low. Nevertheless it will be necessary to determine the appropriate oxidation kinetics so that through life predictions of component oxidation distributions can be determined for different levels of impurities.

The rapid oxidation data derived from the HTR-M/M1 programme showed a good comparable reactivity for most of the selected graphites, with one clear exception (NBG-20) which had a much higher reactivity (see Figure 3-1). The reason for this "unusual" behaviour is not well understood.

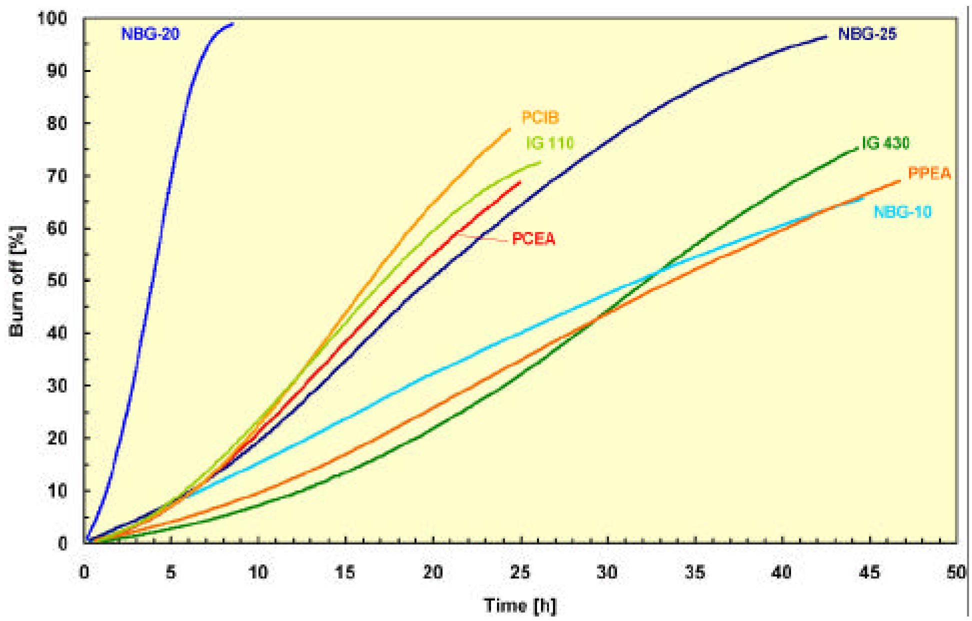

Figure 2-2 Exemplary graph from HTR-M1 for high oxidation rates of the selected graphites at 650 °C in air

3 Design methodologies

3.1 Requirements for calculations

In order to design a graphite core it is necessary to perform the following basic calculations on individual components and the structure as a whole

(i) Calculation of fluence distributions in components

(ii) Calculation of temperature distributions in components

(iii) Calculation of oxidation distributions in components

(iv) Calculation of irradiation induced stress distributions in components

(v) Calculation of irradiation induced distortions in components

(vi) Calculation of loadings generated in the structure

(vii) Calculation of component displacements generated in the structure

Most of the above require special computer codes to be developed, which need to be verified where possible against other codes, and validated where possible against experimental data.

3.2 Existing design codes

There are two draft graphite core design codes in existence, one produced by ASME in the USA and one produced by the KTA in Germany. The ASME code for graphite cores (Ref 6) was produced primarily for the US HTGR programme, but was never progressed further due to the cancellation of the US programme. Around the same time a draft design code for graphite components (Ref 7) was also under development in Germany, again primarily for the country's HTGR programme, and again was never progressed further due to the cancellation of the German programme.

After the construction of the last two AGRs in the UK, a document entitled 'The Compendium of AGR Core and Sleeve Data and Methods' was developed which contains all the graphite data and methods needed to make future operational safety cases for the AGRs. It is a live document and is updated as required to reflect improvements in methodology and revisions to the graphite data. The data sheets cover Young's modulus, strength, dimensional change, coefficient of thermal expansion, thermal conductivity, irradiation induced creep, specific heat and stored energy, gas transport properties (diffusivity ratio and viscous flow coefficients), virgin bulk density and open pore volume. The method sheets cover the calculation of fast neutron fluence, temperature and radiolytic oxidation distributions in components, brick geometry changes, core loads, brick internal stresses and external (load applied) stresses, brick integrity and core geometry.

3.3 ASME code development

An ASME Project Team, initiated in 2004, is currently developing a design code for graphite core components for HTRs. The Project Team currently has members from the USA, UK, France, South Africa and Japan. The existing draft ASME design code was used as the starting point, but as it doesn't adequately cover irradiated components, a major re-write is required. The code when completed will include rules for materials, design, fabrication and testing, including the quality control requirements for fabrication and installation. It is expected that a complete draft of the code will not be available until 2008, and that it will take until at least 2010 before it has undergone a full peer review by ASME and becomes officially incorporated in Section III of the ASME Boiler & Pressure Vessel Code.

3.4 Proposals for European (V)HTR

It is proposed that the graphite data and design methodology for the European (V)HTR be contained in a Compendium similar to that produced by the UK for the AGRs. However, not all the data and methodology sections will be required. The data sections will contain all the necessary data for the grade(s) of graphite selected as being the best overall. The selection will be based on a number of factors including the irradiation behaviour obtained from the FP5, Raphael and subsequent experiments, long-term material availability and cost. Aspects of the methodology contained within the ASME graphite code currently under development will be referenced as required. An outline of the proposed European (V)HTR Compendium is given in Appendix A. It should be noted that methods/computer codes will be required to calculate neutron fluxes (and hence fluences) and temperature distributions within components (the latter depending on the heating and coolant flow rates). These are outside of the scope of the work on materials and will have to be developed by others.

4 Scope and planning

The scope of the work required regarding the development of graphite data and design methodology is as follows:

(i) Determine unirradiated material properties for selected graphites

(ii) Determine irradiation behaviour at specified temperatures over the full fluence range (includes dimensional change, Young's modulus, strength, CTE and thermal conductivity)

(iii) Populate database and produce design curves

(iv) Develop appropriate methodologies for a core design

(v) Produce a compendium of graphite data and design methodology

The planning will have to take into account the timescales for obtaining the necessary irradiation behaviour and also the development of the ASME design code. The timescales for the current irradiation experiments are given in Figure 4.1, which indicates that the data will be available by 2010. However, it will also be necessary to carry out an irradiation creep experiment on selected graphites. Such an experiment is being carried out in the US in three stages, with increasing fluences (dpa, EDND) being reached in successive stages. If the third stage is carried out this should provide all the necessary data, but this is not expected to be completed until well after 2010.

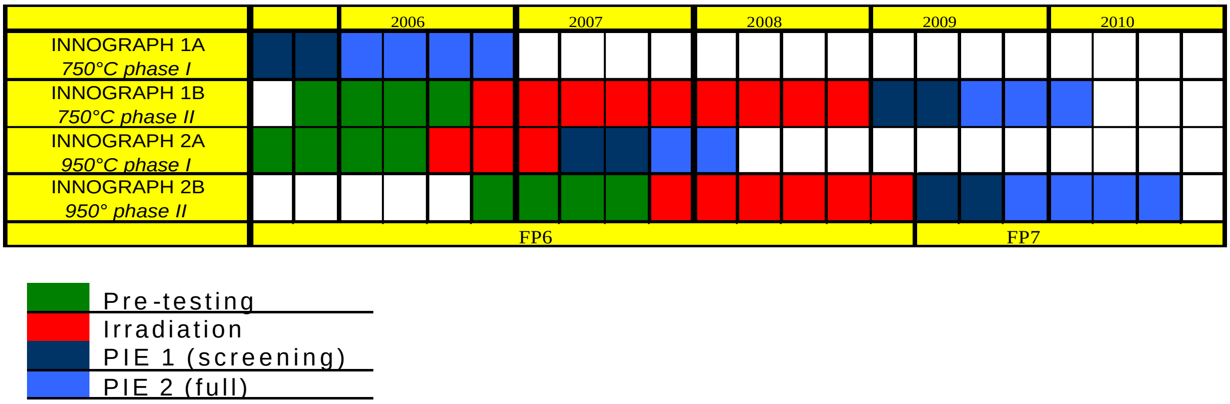

Figure 4-1 Timescale for current irradiation experiments

As mentioned in Section 3.3, it is expected that a complete draft of the ASME graphite code will not be available until 2008, and that it will take until at least 2010 before it has undergone a full peer review by ASME and becomes officially incorporated in Section III of the ASME Boiler & Pressure Vessel Code. This is also the anticipated timescale for completing the development of the design methodology. Nevertheless work on producing the proposed Compendium can start in the near future and the various data and method sub-sections can be drafted and completed as data become available and methods are agreed.

In order to speed the process up and to save time, effort and money by carrying out cost-effective experiments to obtain the necessary data, an agreement for international collaboration is being set up under the Gen IV programme. To facilitate this, a Graphite Working Group (GWG) has been set up under the VHTR Project Management Board on Materials & Components (PMB-M&C). The GWG will have appropriate membership from interested parties, and is actioned to produce and agree a detailed matrix of data requirements for the different graphites. It will identify, and agree on, who should carry out the individual tasks and on what timescale.

5 Conclusion

In order to be able to carry out the initial design of the graphite core in a (Very) High Temperature Reactor (V)HTR it is essential to have reliable graphite data and a proven design methodology. The necessary data on the irradiation behaviour are to be obtained from experiments being carried now and in the future in the HFR at Petten, and the oxidation behaviour from experiments being carried now and in the future at FZJ. It is proposed that graphite data and design methodology for the European (V)HTR be contained in a Compendium similar to that produced by the UK for the AGRs. Aspects of the methodology contained within the ASME graphite code currently under development will be referenced as required. Given the existing European and US irradiation programmes, the complete data set on the irradiation behaviour of the chosen graphites will not be available until well after 2010, although the methodology should be fully developed and agreed by ~2010. Nevertheless, work on producing the proposed compendium can start in the near future.

6 References

Ref	Title
1	J.A. Vreeling, F. Schmalz, D.S. d'Hulst, W. Molijn, P. ten Pierick HTR-M1 Report on preliminary irradiation testing of selected graphites for HTR – report on irradiation and post irradiation tests Report No. HTR-M1 05/12 D 2 3 39
2	M W Davies. Review of the preliminary graphite irradiation results and updating of the graphite database. Report No. HTR-M1 05/12 D 2 4 40
3	M W Davies. Assessment of the preliminary graphite irradiation results from INNOGRAPH 1A and comparison with previous data. Report No. HTR-M1 05/12 D 2 4 41
4	K Kuhn. Comparison of the oxidation behaviour of 8 HTR graphites Report No. HTR-M1 04/12 D 2 0 27
5	HTR-M1 Final Report HTR-M1 05/12 P 0 0 46
6	Proposed Section III, Division 2, Subsection CE, ASME Boiler and Pressure Vessel Code. Design Requirements for Graphite Core Supports.
7	KTA 3232. Design Rule

Appendix 1 – Contents for proposed Data and Methods Compendium

1 Introduction

It is proposed that a Data and Methods Compendium be produced that will provide guidelines on the data and methods to be used for the European (V)HTR graphite core design. It was also proposed that this could similar to the Compendium developed by the UK for the safety assessment of the AGR cores. This Appendix gives an outline of the proposed Compendium for the European (V)HTR.

2 Overall layout

The overall layout of the proposed Compendium is as follows:

(i) Introduction Sheet
(ii) Contents Sheet
(iii) Section 1 - Data Section Cover Sheet plus Data Sheets
(iv) Section 2 - Methods Section Cover Sheet plus Method Sheets

The proposed contents of Section 1 and 2 are covered below.

3 Section 1

Section 1 will contain all the necessary design data for the selected graphite(s). This will included as-manufactured properties and irradiation behaviour covering the full fluence and temperature range. The data will be contained in a number of sub-sections as given below:

(i) Stress–strain relationship (Young's modulus)
(ii) Strength
(iii) Dimensional change behaviour
(iv) Coefficient of thermal expansion
(v) Thermal conductivity
(vi) Irradiation creep
(vii) Oxidation behaviour (rapid + long term)
(viii) Other properties

4 Section 2

Section 2 will contain all the necessary methodology for carrying out a graphite core design for a (V)HTR. A flowchart will be generated that shows the individual methods required and their interrelation to carry out a core design. The methods will be contained in a number of sub-sections as given below. A brief outline of each is given.

4.1 Calculation of fast neutron fluence distributions in a component

In order to be able to determine internal stresses and distortions for individual components (4.5 below) it is necessary to know their fast neutron fluence distributions at any given time in life. There are a number of codes that can be used to do this. In the UK there is MCBEND and WIMSE. Other (international) codes include MCNP.

4.2 Calculation of temperature distributions in a component

In order to be able to determine internal stresses and distortions for individual components it is also necessary to know their through life temperature distributions. There are a number of codes that can be used to do this. In the UK there is the PANTHER code. Other general purpose (international) codes can be used.

4.3 Calculation of core flows and pressure drops

The calculation of core flows is necessary for determining the equilibrium temperatures in components at any time in life (4.3 above). Pressure drops are required to determine the long term oxidation distributions in components (4.4 below) and the external loads on the structure and individual components.

4.4 Calculation of oxidation distributions in a component

Since oxidation reduces the strength of the graphite it is important to be able calculate the long-term oxidation distribution in components due to impurities in the coolant gas, and also the rapid oxidation that can arise due to air ingress accidents. This requires the development of the appropriate code although it is possible that some codes already exist (for earlier HTR designs) which could be used as a starting point.

4.5 Calculation of component internal stresses and distortions

It is extremely important to now how the stresses in a component due to the dimensional change behaviour are developing with time and also the additional transient thermal stresses that arise during shut-down to cold conditions. These can be much larger than the stresses due to external loading and will eventually lead to cracking, particularly when the strength of the material begins to reduce. A number of computer codes do exist but these have been tailored to the particular graphites (and perhaps core designs) used in each country. There is a move within the VHTR Gen IV programme to unify the methodology.

4.6 Calculation of interaction with interfacing structures

The graphite core will almost certainly be supported by a steel support system and be radially constrained by some type of restraint system. During normal operation such as reactor start-up and shut-down, and particularly during fault conditions, the differential movement between the structures can lead to interactions between them which can induce changes to core geometry and additional loading on components. It is therefore important to be able to calculate the interaction between the core and interfacing structures. As for the stress analysis (4.5 above) a number of computer codes do exist but these have been tailored to the particular reactor designs used in each country. It will not be possible to develop such a code until the concept design has been decided, although the recommended methodology can be specified e.g. using Finite Element (FE) solid modelling.

4.7 Calculation of external loads

External loads on the core arise due dead-weight, component distortions, and the interaction between the core and interfacing structures. Again, a number of computer codes do exist but these have been tailored to the particular reactor designs used in each country. It will not be possible to develop such codes until the concept design has been decided, although the recommended methodology can be specified e.g. using FE solid modelling

4.8 Calculation of applied stresses from external loads

Once the external loads on individual components are known it will be necessary to determine the stresses in each. Other than dead weight loads, most other loads will be transmitted through the interconnecting features used such as keys and keyways, and dowels. The stresses can be calculated using conventional FE modelling techniques. In order to achieve consistency between the different calculated stresses it is important to use the same mesh refinement for each.

4.9 Calculation of core movements (channel geometry)

One of the fundamental design requirements for the core will be that it will provide the required constraint on fuel and unimpeded access for control rods and other devices. It is therefore important to be able to predict the changes in geometry that can arise due to the predicted component distortions and other influences.

4.10 Calculation of component integrity

Having made predictions of how the internal stresses with components vary with core life and the magnitude of the transient thermal stresses and the external (load applied) stresses, it is important to combine these in the appropriate way and to compare them with a failure stress in order to determine the likelihood of a crack initiating, which if propagates could lead to a loss of component functionality.

Note - The capability of a core design to operate safely with cracked graphite components will depend on a number of factors and its demonstration is likely to involve complex modelling substantiated by experimental rig testing. This was the case for the AGRs. Such core states will not be covered by the ASME design code and so it is not proposed to include this in the compendium.