



EUROPEAN COMMISSION
5th EURATOM FRAMEWORK PROGRAMME 1998-2002
KEY ACTION : NUCLEAR FISSION

HTR-M

**European Project for the development of HTR Technology -
Materials for the HTR**

CONTRACT N°
FIKI-CT-2000-00032
FIKI-CT-2001-00135

**Review of the preliminary graphite irradiation results
and updating of database**

by

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U.K.

Dissemination level : CO
Document Number: HTR-M1 05/12 D 2 4 40
Deliverable D10

Commercial-in-Confidence

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**C6857/TR/005
Issue 01**

Commercial-in-Confidence

DOCUMENT ISSUE RECORD

(engineering documents)



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Document Title : Review of the preliminary graphite irradiation results and updating of the graphite database

Project Reference: C6857

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				Dec 2005
		M W Davies	D E Buckthorpe		
01	Checker and partner comments incorporated	<i>M W Davies</i>	<i>D E Buckthorpe</i>	<i>T Lennox</i>	
		M W Davies	D E Buckthorpe	T Lennox	

Total number of pages:

Intro:

(i)-(iv)

Text

1 - 6

Tables

T1-T3

Figures

Appendices

Previous issues of this document shall be destroyed or marked **SUPERSEDED**

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

3050aJan01

Controlling procedure - QP11, QP40

Docprodref

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Summary

The first stage of an irradiation programme has been carried out in the Petten HFR involving ~200 samples manufactured from eight different grades of graphite. This is referred to as INNOGRAPH1A. 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

For the initial pre-characterisation of the samples their initial dimensions, mass, dynamic Young's modulus (DYM), coefficient of thermal expansion (CTE) and thermal diffusivity were measured. (The latter is used to calculate thermal conductivity.) From these, their volumes and densities were calculated. Around 50% of the irradiated samples have been subjected to some PIE, having their dimensions, mass and DYM re-measured.

All the pre-characterisation measurements, and the PIE measurements made to date (together with specific properties derived from them) are contained within Excel spreadsheets developed by NRG. These have now been transferred to individual worksheets for each graphite grade. These worksheets are used to calculate the fractional changes in the properties due to irradiation and to display them in graphical form. Eventually, it is intended that they be used to produce the appropriate 'design curves'.

Further upgrades will be carried out whenever measurements on samples are carried out. In the short term these will include further PIE measurements on the irradiated samples from the INNOGRAPH1A irradiation, and the pre-characterisation of the samples for the INNOGRAPH1B and INNOGRAPH2A irradiations.

1 Introduction

In order to support the development of a future European High Temperature Reactor (HTR), a number of suitable candidate graphites were selected for an irradiation programme in the high flux reactor (HFR) at Petten, which is being undertaken by NRG. The overall irradiation programme is aimed at establishing the irradiation behaviour of the selected graphites over the irradiation fluence and temperature range appropriate for a HTR, and eventually to provide all the graphite data necessary to undertake a core design. (Details on the selected graphites and testing requirements are given in Refs 1 and 2).

The first stage of this irradiation programme has been carried out within the 5th Framework Programme (HTR-M1). Appropriately sized discs/blocks of the different materials were supplied to NRG by three different graphite manufacturers and ~200 samples were machined from them. The samples were first of all pre-characterised, and then irradiated to a maximum of around 1/3 peak fluence¹ at a temperature of 750°C. This is referred to as INNOGRAPH 1A. [The peak fluence to be achieved is ~25 displacements per atom (dpa), or $\sim 200 \times 10^{20} \text{ n.cm}^{-2}$ Equivalent DIDO Nickel Dose (EDND). Thus in INNOGRAPH 1A the samples have been irradiated up to ~9 dpa, or $\sim 70 \times 10^{20} \text{ n.cm}^{-2}$ EDND. The irradiation to peak fluence will be completed in INNOGRAPH 1B]. 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).

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). From these, their volumes and densities were calculated. For the irradiated samples subjected to PIE, only their dimensions and DYM have been re-measured to date.

All the pre-characterisation measurements and the PIE measurements already made (together with specific properties derived from them) are contained within Excel spreadsheets developed by NRG. However, 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 produced (Ref 3). At the conclusion of the HTR-M1 programme there was a requirement to review the available measurements and to enter them into this worksheet. This is the subject of this report. (There was also a requirement to assess the measurements and compare them with those made on other graphites in the past at similar temperatures. A preliminary comparison has been carried out and this is reported separately in Ref 4.)

A summary of the graphites tested is given in Section 2. (Full details of the selection process and the graphites selected are given in Ref 1 and 2). The pre-characterisation of the graphites is covered in Section 3 and the PIE measurements in Section 4. The dimensional change behaviour and the calculated variations in DYM are covered in Section 5, and future work to be undertaken is covered in Section 6.

¹ Peak fluence is that appropriate to a pebble bed core design. The replaceable components in a prismatic core design should not exceed 1/3 of this value and thus the data requirements should be bounded by this first stage of the irradiation.

2 Graphite selection

There is clearly a limit on the number of samples that can be contained within a given irradiation experiment and this has an influence on the number of graphites that can be tested. There is also a need to maximise the number of samples of each graphite selected to provide good statistics. One problem was that the general anisotropy of the different graphites requires samples to be machined from the two principal directions, that is, 'with grain' (WG) and 'against grain' (AG). In addition, due to the inherent material variability within a block, samples should be taken from both the centre region of a block and from the outside region of the block (hereafter referred to as 'centre' and 'edge' in this report).

After careful consideration eight graphites were chosen from those available for this initial experiment, five of which were classified as 'major' grades, the other three being classified as 'minor' grades. The intention was to cover the two main sources of coke (petroleum and pitch), and the two most common manufacturing techniques (extrusion and iso-moulding). Due to a decision to use a mixture of two sample lengths (6 mm and 12 mm) it was possible to increase the number of samples that could be tested to 198. Therefore, in general, 30 samples of each were produced for the five 'major' grades (15 in each of the two principal directions), and 14 samples of each were to be produced for the 'minor' grades (7 in each of the two principal directions).

The selected major and minor graphites are shown in the Table below. (The actual number of samples of each included is given in parentheses after the classification.) All are medium grain graphites, with the exception of PCIB which is a super-fine grain graphite.

MANUFACTURER	GRADE	COKE	PROCESS	CLASSIFICATION
GrafTech	PCEA	Petroleum	Extrusion	Major (30)
	PPEA	Pitch	Extrusion	Minor (14)
	PCIB	Petroleum	Iso-moulding	Major (30)
SGL	NBG-10	Pitch	Extrusion	Major (32)
	NBG-20	Petroleum	Extrusion	Minor (14)
	NBG-25	Petroleum	Iso-moulding	Major (30)
Toyo Tanso	IG-110	Petroleum	Iso-moulding	Major (30)
	IG-430	Pitch	Iso-moulding	Minor (18)

3 Pre-characterisation measurements

The graphite discs/blocks supplied by the manufacturers for each grade were sectioned and machined to produce the required number of samples. Each sample was cylindrically shaped with a nominal diameter of 8 mm and a nominal length of 6 or 12 mm. As mentioned earlier, the samples were machined from both the central and outer regions of each block, to take account of the known variability in properties throughout a block. An additional small flat was also machined so that each could be uniquely identified with an alphanumeric code, and its orientation within the block known for recording the measurements (Ref 5). In this way, initial property variation

as a function of position and orientation in the disc/block could be determined (as well as for the irradiated properties).

The dimensions of each sample were measured together with its DYM, CTE and thermal diffusivity (used to calculate thermal conductivity). The mass of each sample was also measured, which together with the dimensions, allowed the initial density to be determined. Details of the testing methods used are given in Ref 6. (Although strength is a very important property, it cannot be measured on the samples to be irradiated as this will involve a destructive test. Nevertheless, strength measurements can be made on neighbouring samples obtained from a block, and the strength measurements thus obtained should be representative of those being irradiated.) All the measurements have been recorded in the spreadsheet developed by NRG.

The initial task for this report was to transfer the measurements from the NRG spreadsheet into the proposed worksheet. This would then allow the property variations to be displayed in graphical form and the appropriate 'design curves' fitted. However, it soon became apparent that a single worksheet for all eight graphites would be too large and difficult to use and so it was decided that individual worksheets should be produced for each grade. There was also a need to amend each basic spreadsheet slightly in order to facilitate data entry and the generation of graphs/curves from them.

Worksheets for all eight graphites have been produced and the spreadsheet within each has been populated with the pre-characterisation measurement data. This spreadsheet is split into three 'pages'. The first 'page' contains the pre-characterisation data, the second 'page' contains the PIE measurements, and the third 'page' contains the calculated fractional changes in the properties. Table 1 is an example of the first page of such a spreadsheet, and shows the pre-characterisation measurements for IG-110.

It can be seen that a total of 30 samples were pre-characterised, 16 of which were from the centre of the block and the other 14 from the edge of the block. Of the 16 samples from the centre of the block, 8 were machined with the cylindrical axis in the AG direction and the remaining 8 were machined with the cylindrical axis in the WG direction. Of the 14 samples from the edge of the block, 7 were machined with the cylindrical axis in the AG direction and the remaining 7 were machined with the cylindrical axis in the WG direction.

Measurements were made of the initial length (L_0) and diameter (D_0) of each specimen, together with the shorter diameter perpendicular to the machined flat (x_0). From these the initial volume (V_0) of the specimen was calculated. The initial mass (M_0) was measured and from this and V_0 the initial density (ρ_0) was calculated. The DYM was measured along the axis only ('L' direction). The CTE of the specimen was also only measured along its axis, but over three temperature ranges (30 – 120°), (30 – 200°), and (30 – 750°). The value quoted for each is the average over the range. The thermal diffusivity (a) was measured at 27°C and at 100°C intervals between 100°C and 700°C. From each of these measurements the thermal conductivity (λ) was calculated using the formula $\lambda = a r C_p$ where r is the density and C_p is the specific heat of the specimen.

Due to the length of time required to carry out individual CTE and thermal diffusivity measurements it was not practical to carry out these measurements on all the samples. Therefore samples that were close neighbours to those that were measured were allocated the same values.

4 Post-irradiation measurements

All the measurements carried out for the pre-characterisation are also required on the irradiated samples. At the time of writing, a total of 96 samples have been subject to measurement (~50%), but to date, it has only been possible to measure their linear dimensions, mass and DYM, and to calculate (from the first two) their volumes and density.

The second 'page' of the spreadsheet within each of the worksheets has been populated with the PIE measurement data. Table 2 is an example of such a 'page', and shows the PIE measurements for IG-110. As well as the measurements, it is also important to know the fluence seen by each sample. This is expressed in the Table in two ways, firstly in terms of displacements per atom (dpa) and secondly in terms of Equivalent DIDO Nickel Dose (EDND). It can be seen that the fluence reached ranges from 6.85 to 9.84 dpa, or 53.9 to 77.4 in terms of EDND (10^{20} n.cm⁻²). The mid range is 8.35 dpa (65.7×10^{20} n.cm⁻² EDND), which is 1/3 of the target full fluence of 25 dpa (200×10^{20} n.cm⁻² EDND),

5 Changes in properties due to irradiation

It is known that the properties of interest to the core designer all vary with irradiation fluence and that this variation also depends on the irradiation temperature. The linear dimensional and volumetric changes that occur are traditionally given in terms of fractional changes, expressed as a percentage. The variation is normally expressed as a ratio so that it can be used as a multiplying factor (f) for the initial modulus (E_0) to give the irradiated modulus (E_i) at the required fluence/temperature. In order to calculate the changes, the pre-characterisation measurements and the post-irradiation measurements are entered into the appropriate locations the first and second 'pages' of the spreadsheet respectively. The variations are calculated in the required manner in the third 'page' of the spreadsheet.

Table 3 is an example of such a 'page', and shows the calculated changes for IG-110. This is the 'page' used to generate the graphical plots and is therefore made 'stand-alone' by also providing information on the location and orientation of the samples as well as the fluence. (The production of the graphical plots and the assessment of the behaviour of the different graphites is carried out in Ref 4)

6 Future work

The remaining measurements on the samples subjected to PIE to date, and all the measurements on the remaining samples, will be carried out over the next 6-9 months. The worksheets will be updated again when all the data are available.

The second stage of this irradiation (to full fluence) will be undertaken shortly. Around 1/2 of the samples will be replaced by new samples of the selected graphites.

These will therefore be taken to ~2/3 full fluence. The new samples are being pre-characterised and the data will be added to the worksheets when available.

A similar irradiation at 950°C is to be carried out, initially to 1/3 full fluence, in the 6th Framework Programme (Raphael). Pre-characterisation measurements on the prepared samples will be available during 2006. The worksheets will be updated to incorporate these data. Additional worksheets will be generated for each the three new graphites to be tested (NBG-18 as a major grade, and NBG-17 and BAN as minor grades)

7 Conclusions

The first stage of an irradiation programme has been carried out in the Petten HFR involving ~200 samples manufactured from eight different grades of graphite. 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

For the initial pre-characterisation of the samples their initial dimensions, mass, dynamic Young's modulus (DYM), coefficient of thermal expansion (CTE) and thermal diffusivity were measured. (The latter is used to calculate thermal conductivity.) From these, their volumes and densities were calculated. Around 50% of the irradiated samples have been subjected to some PIE, having their dimensions, mass and DYM re-measured.

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8 References

Ref	Title
1	M W Davies. Report on selection of graphites and testing requirements. HTR-M1-02/11-D-2.1.9. NNC Report C6857/TR/001
2	M W Davies. Assessment of new potential graphites and needs for HTR. HTR-M-02/11-D-3.2.53. NNC Report C6463/TR/019
3	M W Davies. Report on database of graphite properties for use by designers. HTR-M-02/06-D-3.1.22. NNC Report C6463/TR/012.

Ref	Title
4	M W Davies. Assessment of the preliminary graphite irradiation results from INNOGRAPH1A and comparison with previous data. HTR-M1-05/12-D-2.4.41. NNC Report C6857/TR/006
5	F Schmalz et al. Preliminary irradiation testing of selected graphite grades for HTR. HTR-M1-05/12 - D-2.3.39
6	A Vreeling. HTR-M1 report on specimen fabrication and procurement for the graphite irradiation tests. HTR-M1-03/04-D-2.2.13

Table 1 Pre-characterisation measurements for IG-110

[illegible]

Table 2 Post-irradiation measurements for IG-110

Graphite materials property database										Graphite:		IG-110														
										Manufacturer:		Toyo tanso														
										Coke:		Petroleum														
										Grain size:		Fine														
										Process:		Iso-moulding														
										Irradiated properties																
Irrad temp (°C)	Fluence		Length L _i (mm)	Diam D _i (mm)	Diam x _i (mm)	Vol V _i (mm ³)	Mass M _i (g)	Density ρ _i (g/cm ³)	DYM E _i (GPa)	CTE α _i (x10 ⁻⁶ /K)					Thermal conductivity (W/m/K)											
	DPA	EDND (10E20)								30-120	30-750	30-200	27°C	100°C	200°C	300°C	400°C	500°C	600°C	700°C						
750	7.73	58.7	11.7906	7.8506	7.6296	566.19	1.04038	1.8375	21.40																	
750																										
750																										
750	6.85	52.1	5.9104	7.8778	7.6288	285.36	0.51834	1.8164	18.54																	
750	7.71	58.6	5.8713	7.8568	7.6065	281.93	0.51211	1.8164	18.14																	
750	9.84	74.8	5.8699	7.8066	7.5738	278.53	0.51242	1.8398	22.85																	
750																										
750	9.68	73.6	5.9645	7.816	7.5689	283.47	0.5321	1.8771	24.60																	
750	6.68	50.8	5.8438	7.8927	7.5714	281.98	0.51638	1.8313	20.63																	
750	9.10	69.2	5.8202	7.8757	7.5514	279.56	0.52003	1.8602	27.87																	
750																										
750	9.73	73.9	5.8344	7.8708	7.5936	280.72	0.52674	1.8764	28.95																	
750																										
750	8.60	65.4	11.8466	7.8731	7.613	570.91	1.05766	1.8526	24.91																	
750																										
750																										
750																										
750																										
750	7.99	60.7	5.8855	7.8547	7.6082	282.52	0.52292	1.8509	22.05																	
750	9.09	69.1	5.9687	7.8233	7.5965	284.53	0.52905	1.8594	23.90																	
750																										
750																										
750																										
750	7.87	59.8	5.8473	7.8494	7.6055	280.34	0.51847	1.8494	24.42																	
750	9.12	69.3	5.7605	7.8768	7.6142	277.94	0.51723	1.8609	26.36																	
750																										

Table 3 Property changes for IG-110

Graphite materials property database										Graphite: IG-110		Fractional change													
Location	Orientation	Fluence		Length L (%)	Diam D (%)	Diam x (%)	Vol V (%)	Mass M	Density ρ	DYM E (GPa)	CTE						Thermal conductivity								
		DPA	EDND (10E20)								30-120	30-750	30-200	27°C	100°C	200°C	300°C	400°C	500°C	600°C	700°C				
CENT	AG	7.73	58.7	-1.1478	-1.5932	-1.8941	-4.3905	1.9E-05	0.04594	2.33575															
CENT	AG																								
CENT	AG																								
CENT	AG	6.85	52.1	-0.9646	-1.3619	-1.166	-3.5593	-0.0011	0.03576	2.04624															
CENT	AG	7.71	58.6	-1.0461	-1.6123	-1.5807	-4.1982	-0.0011	0.04266	2.07835															
CENT	AG	9.84	74.8	-0.8158	-2.2105	-1.8459	-4.997	-2E-16	0.05264	2.61332															
CENT	AG																								
CENT	AG	9.68	73.6	-0.797	-2.1605	-1.9009	-4.9259	0.01625	0.06891	2.78274															
CENT	WG	6.68	50.8	-1.4279	-1.1235	-0.781	-3.4631	-0.0009	0.03495	1.96152															
CENT	WG	9.10	69.2	-2.156	-1.3611	-1.1014	-4.6773	9E-05	0.04918	2.57512															
CENT	WG																								
CENT	WG	9.73	73.9	-2.404	-1.4232	-2.1288	-5.4594	-0.0002	0.05755	2.59676															
CENT	WG																								
CENT	WG	8.60	65.4	-1.8747	-1.5096	-1.2585	-4.7058	-0.0007	0.04864	2.19934															
CENT	WG																								
CENT	WG																								
CENT	AG																								
EDGE	AG																								
EDGE	AG	7.99	60.7	-1.1989	-1.7255	-1.4303	-4.4531	-0.0002	0.04643	2.34635															
EDGE	AG	9.09	69.1	-1.2508	-1.9553	-1.4926	-4.8806	-0.0007	0.05053	2.56371															
EDGE	AG																								
EDGE	AG																								
EDGE	AG																								
EDGE	WG	7.87	59.8	-2.0525	-1.2579	-1.0851	-4.4312	-0.0004	0.04591	2.27349															
EDGE	WG	9.12	69.3	-2.2739	-1.4076	-1.1853	-4.9093	-0.0014	0.0501	2.48352															
EDGE	WG																								