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The need for a graphite heat-up experiment

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The need for a graphite heat-up experiment

Abstract

When subject to fast neutron irradiation, graphite exhibits dimensional change and almost every property is affected. One such property is thermal conductivity, which falls quite dramatically. This could significantly affect the ability to conduct heat out of the core during a fault condition, and thus the peak fuel temperature. It is known however that the irradiated thermal conductivity partially recovers if the graphite temperature is increased above that at which it was irradiated. It might be necessary to claim this partial recovery when making a safety case for the reactor for such a fault condition. There is therefore a need to determine the amount and rate of recovery of thermal conductivity against temperature. This report outlines the proposed experiment to achieve this.

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1 Introduction

It is known that, when subject to fast neutron irradiation, graphite exhibits dimensional change and almost every property is affected. One such property is thermal conductivity, which falls quite dramatically. This could significantly affect the ability to conduct heat out of the core during a fault condition, and thus the peak fuel temperature. It is known however that the thermal conductivity of irradiated graphite partially recovers if the temperature is increased above that at which it was irradiated. It might be necessary to claim this partial recovery when making a safety case for the reactor for such a fault condition. There is therefore a need to determine the amount and rate of recovery of thermal conductivity against temperature.

This report outlines the proposed experiment to achieve this. Section 2 lists the selected graphites and gives details of the planned irradiation experiments. Section 3 illustrates the typical variation in thermal conductivity of a nuclear grade graphite with irradiation fluence and temperature, explains the recovery with increasing temperature and outlines the extent of testing required to determine this for the selected graphites. Section 4 describes the planned experimental set-up to obtain the data.

2 Selected graphites and planned irradiation experiments

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 Planned irradiation experiments

Irradiation experiments are required to provide data up to the peak fluence expected and over the appropriate temperature range. 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). 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. Thus in INNOGRAPH 1A the samples have been irradiated up to ~9 dpa, or ~70 x 10^{20} n.cm^{-2} EDND. The test rig has been removed and dismantled and, at the time of writing of this report, most of the samples have already been subjected to full post-irradiation examination (PIE). The irradiation to peak fluence will be completed in INNOGRAPH 1B, which is about to commence.

A similar irradiation at 950°C is to be carried out, initially to 1/3 peak fluence, in FP6 (Raphael). This is referred to as INNOGRAPH 2A. This has recently commenced and will be completed in 2007. The second irradiation stage to peak fluence at this temperature (referred to as INNOGRAPH 2B) will be undertaken between late 2007 and early 2009 but the full set of PIE measurements on the samples at the end of this irradiation will not be available until ~2010. Table 2-2 gives the timescale for, and Figure 2-1 gives a schematic of the data ranges to be expected from, the different irradiation experiments within HTR-M1 and Raphael-IP.

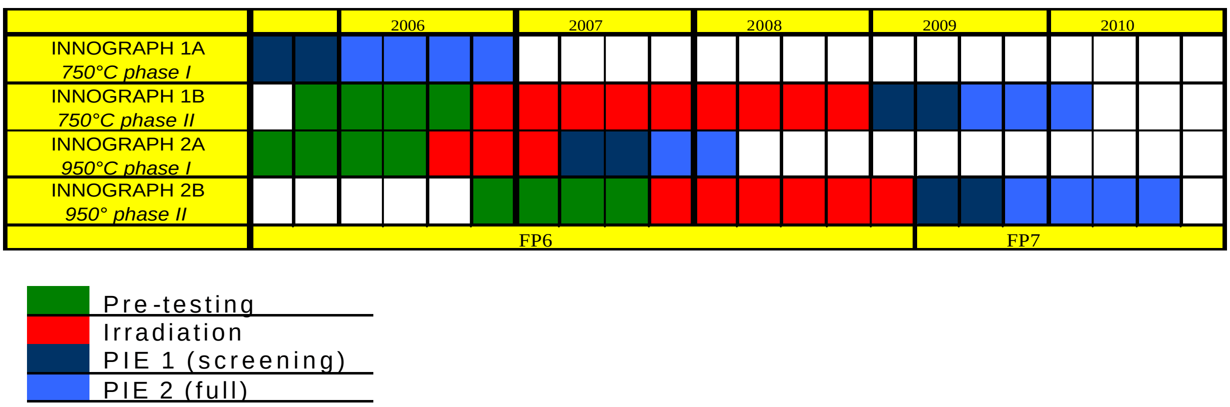

Table 2-2 Timescale for current irradiation experiments

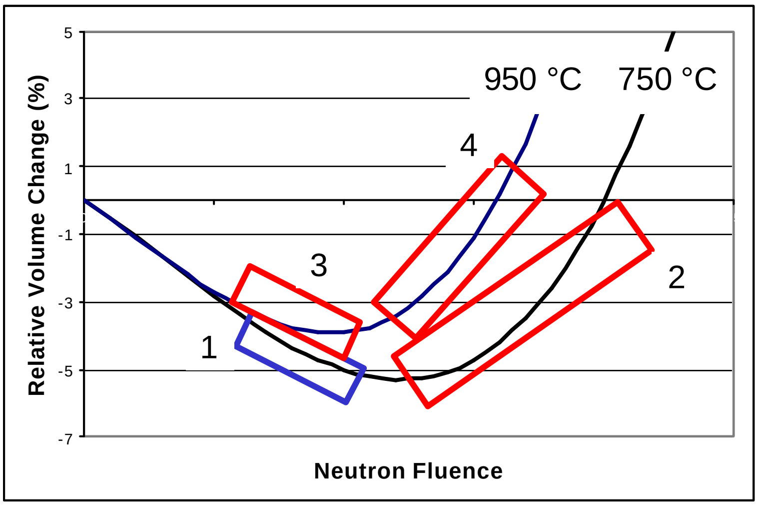

Figure 2-1 Schematic overview of the different irradiation experiments within HTR-M1 and Raphael-IP

3 Thermal conductivity changes

3.1 Typical variation with irradiation and temperature

As stated earlier the thermal conductivity of nuclear grade graphite drops quite markedly when subject to fast neutron irradiation. This is illustrated in Figures 3.1. The unirradiated value is typically about 130 W/m K. After a relatively small fluence, this drops to around 20-30 W/m K, i.e. by a factor of about 5 to 6. It remains fairly constant thereafter until a much higher fluence is reached when it falls further.

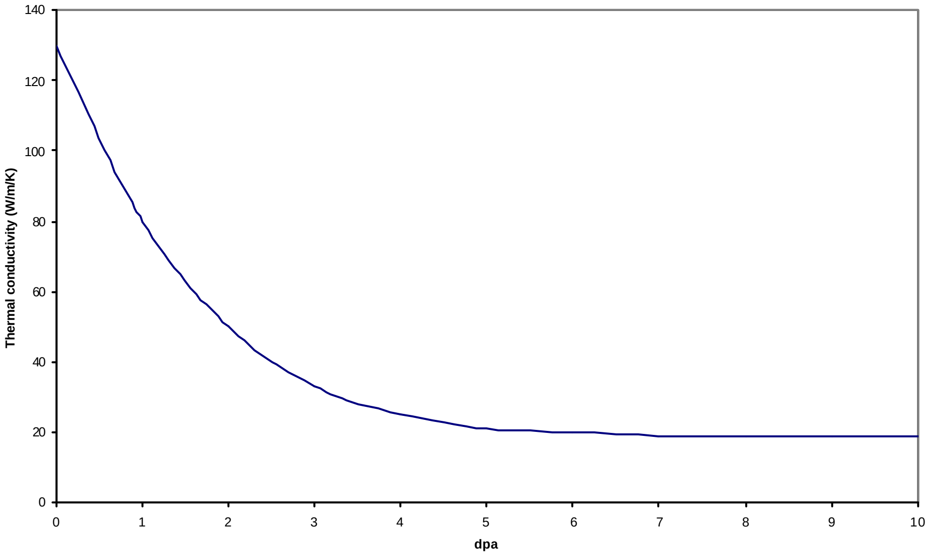

Figure 3.1 Typical thermal conductivity variation with irradiation

Thermal conductivity/diffusivity also varies with temperature as illustrated in Figure 3-2 below for NBG 10. It can be seen that the unirradiated diffusivity falls at a decreasing rate from around 110 mm^2/s at 20^{o}C to around 22 mm^2/s at 700^{o}C i.e. by a factor of 5. The irradiated value at 20^{o}C is only around 22 mm^2/s (i.e. around one fifth of the unirradiated value) and this falls at a decreasing rate to around 11mm^2/s at 700^{o}C i.e. only by a factor of 2. (Note – the peak temperature reached is just below the irradiation temperature and so the recovery with increased temperature has not yet started.)

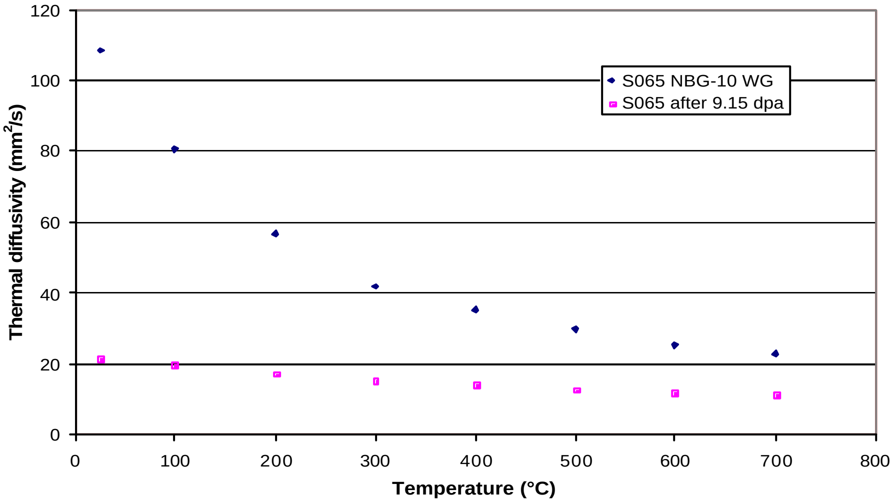

Figure 3-2 Thermal diffusivity variation with temperature for NBG 10 before and after irradiation

3.2 Recovery with temperature

It is known that the thermal conductivity of irradiate graphite recovers to a certain extent when its temperature is increased above that at which it was irradiated. The amount recovered and the rate at which this happens may depend on the irradiation temperature and graphite grade as well as the annealing temperature and its duration. For the graphites irradiated in INNOGRAPH 1A it would be expected that recovery would start when the temperature exceeds ~750^{o}C.

3.3 Extent of testing required

The samples available at the present time are those that have been irradiated in INNOGRAPH 1A and are not being reinstated in INNOGRAPH 1B. There is a range of irradiation fluences that can be tested, but most will have been irradiated at 750^{o}C (with a few at ~670^{o}C). These fluences (6 to 10 dpa) will be large enough for the thermal conductivity of each to have fallen to the initial low value (as illustrated by Figure 3-1), and therefore all the samples should be suitable for determining the recovery of thermal conductivity with temperature. A selection will be made from these. The next set of samples that will become available will be from INNOGRAPH 2A (in 2007). These will also have seen fluences large enough for the thermal conductivity of each to have fallen to the initial low value and thus data on thermal conductivity recovery for graphite irradiated at 950^{o}C can be obtained. In this case it would be expected that recovery would start when the temperature exceeds ~950^{o}C

Samples that have seen ~2/3 and ~full peak fluence will not be available until ~2009. At these fluences, the further expected reduction in thermal conductivity may have arisen. Further experiments may need to be carried out on selected samples as well in order to complete the design data base.

4 Effect on graphite (and hence fuel) temperatures

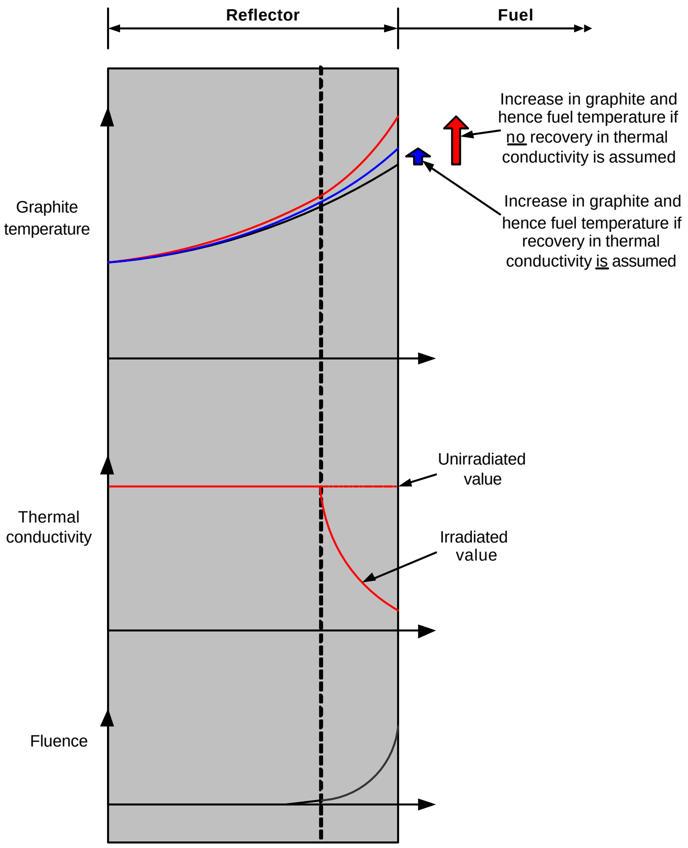

Figure 4-1 Illustration of fluence, thermal conductivity and graphite temperature distributions in the side reflector

The figure above illustrates the fast neutron fluence variation through the side reflector of an HTR and the depth at which it is negligible from the point of view of thermal conductivity changes. The through reflector variation in thermal conductivity is shown above the fluence variation. It can be seen that a large part of the reflector is unaffected by the irradiation, but there is still a significant proportion that is affected.

The figure also shows an illustration of the consequences of thermal conductivity changes with irradiation on calculated graphite temperatures. No effect of irradiation on the graphite thermal conductivity yields the lowest temperature distribution (black line), assuming no recovery in thermal conductivity with temperature gives the highest temperature (red line), and if the recovery in thermal conductivity with temperature is included then the temperature distribution is between the two (blue line). Also since the reflector will be conducting less heat away, the case with no thermal recovery included gives comparatively higher peak fuel temperatures. It is possible that in certain situations with no recovery included the calculated fuel temperatures may be too high to make a safety case.

5 Experimental set-up

The experimental facilities foreseen to be used are:

(i) For simulation of temperature transients:

- The available dilatometry devices (Netzsch 402C) will allow the application of temperature cycles in the range of 1500-1600 ºC. For multiple days at high temperature it is preferred to restrict it to 1550 ºC. Heating rates can be chosen from 1 ºC/min, which is considered of minor importance.

(ii) For quantification of property changes:

- Thermal conductivity through laser flash diffusivity measurements, using Netzsch LFA 457 Microflash. This equipment allows thermal diffusivity measurements up to 1100 ºC.
- Thermal expansion through the mentioned dilatometry devices (Netzsch 402C).
- DYM by time-of-flight measurements as described in [Ref 1]

The experiments will be carried out by NRG. It is planned to determine the extent and are of recovery of thermal conductivity for selected annealing scenarios. The selection of samples will be made from the major grades irradiated in INNOGRAPH 1A.

6 Conclusions

It is known that the thermal conductivity of graphite, when irradiated, falls quite dramatically. This could significantly affect the ability to conduct heat out of the core during a fault condition, and thus the peak fuel temperature. It is known however that the thermal conductivity of irradiated graphite partially recovers if the temperature is increased above that at which it was irradiated. It might be necessary to claim this partial recovery when making a safety case for the reactor for such a fault condition.

It is therefore proposed that a set of experiments be carried out to determine the extent and rate of recovery of the thermal conductivity with temperature. Samples for testing will be selected from the major grade graphites irradiated in the INNOGRAPH 1A experiment. The experiments will be carried out by NRG using existing facilities

7 References

Ref	Title
1	O Wouters, A Vreeling: INNOGRAPH-1B and INNOGRAPH-2A specimen manufacture and pre-characterization -*Graphite material procurement & characterisation* - . Report No. RAPHAEL 0619 D-ML3.3