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Results from EU 5th Framework HTR projects
HTR-M & HTR-M1

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Results from EU 5th Framework HTR projects HTR-M & HTR-M1

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Abstract – The High Temperature Reactor (HTR) projects HTR-M & HTR-M1 have been launched within the 5th Framework Programme (5FP) of the European Commission to consolidate and advance HTR technology in Europe. The projects investigate materials selection and requirements for three key component areas of the HTR: namely the reactor pressure vessel (RPV), the high temperature resistant alloys for the internal structures and turbine, and the graphite for the reactor core. The programme of work includes irradiation testing of vessel steel and graphite materials plus tests on effects of corrosion/ oxidation to determine the effect of HTR environment:

- For the RPV material, fabrication and the effects of irradiation/environment on the HTR vessel weld behaviour are crucial issues with respect to structural viability. Results from investigations and tests carried out to-date are discussed in light of qualification requirements. Recommendations are made for future investigations and tests including creep and creep/fatigue.
- For the High Temperature materials including the turbine, temperature levels and environmental degradation are the main concerns for the direct cycle environment. Fabrication, creep and the effect of helium on properties and strength are assessed for the materials investigated. Fabrication and effects of irradiation/environment are crucial issues for C-based materials seen as future candidates for control rods. Recommendations are made for future investigations and tests and for other component applications including heat exchangers.
- For the graphite materials**:** the investigation of properties under irradiation and corrosion are seen as crucial and an important first step in qualifying new materials for future HTR's given that almost all the graphites previously irradiated are no longer manufactured. Recommendations are made with respect to future test programmes and a graphite selection strategy for HTR application.

The main results obtained to date are discussed and presented in the paper

1. Introduction

HTR-M & M1 projects have been launched within the 5th Framework Programme (5FP) to consolidate and advance HTR materials technology in Europe and are part of a cluster of research projects [1] being performed to investigate research and development issues connected with High and Very High Temperature Reactors (HTR's & VHTR's).

The HTR-M project began towards the end of 2000 and the HTR-M1 project in the autumn of 2001 and jointly they involve eight partners from five different European countries [2]. Within the HTR-M & M1 projects work has been performed to highlight the materials requirements for the HTR reactor pressure vessel, high temperature resistant alloys for the internal structures and turbine, and graphite for the reactor core. The HTR-M & HTR-M1 projects jointly consider two main areas of investigation: the review of past experience and assembly of available properties (including available design information) plus tests on key materials where necessary information is considered lacking or scarce. The HTR-M project focuses primarily on vessel and turbine materials activities and the HTR-M1 project on intermediate creep testing of turbine materials plus irradiation testing of graphites. The technical tasks addressed by the projects are considered to be at the forefront of understanding of materials behaviour and the results are expected to have an important impact on materials selection and development for future HTR and VHTR modular designs.

2. Programme Tasks

The need for reliable material property data is a key issue in the development of the HTR and VHTR technology. The development of advanced HTR concepts requires an understanding of material behaviour plus data information under representative reactor operating conditions for the main HTR components important to safety and feasibility.

The combined programme covers three main areas of work:

Reactor Pressure Vessel (RPV):

- Review existing materials used in gas-cooled reactors and former high temperature reactors plus materials database on design properties (which will lead to identification of data omissions for future R&D testing)
- Carry out specific tests on welded joints under irradiated and non-irradiated conditions.

Fabrication considerations and the effects of irradiation/environment on the vessel weld behaviour are considered crucial issues in determining the structural viability of the material for future RPV application.

The project resources allow one type of vessel steel to be irradiated and a recommendation was made, depending on operating conditions, data omissions, available property and fabrication details.

High Temperature Materials – (Control Rod plus turbine disc & blade):

- Review existing materials used in gas-cooled reactors and previous high temperature reactors plus materials database on design properties
- Perform specific tests on selected materials at temperature and under short times in air, and simulated environments.

The main concern is the ability of the material to withstand the temperatures and conditions in the direct cycle environment. Creep and effects of helium on properties and strength are crucial issues in the selection of suitable materials.

Graphite core:

- Review of existing data and preliminaries for a database plus identification of currently manufactured graphites suitable for HTR core application
- Graphite oxidation and the investigation of the consequences of severe air ingress with core burning, including the development of models and data, protective coatings and C-based materials.
- Perform tests under irradiated conditions on selected materials

The graphite programme is an important first step in establishing suitable materials for future HTR's given that almost all the graphites previously irradiated are no longer manufactured. Note that of the test results that do exist, most use irradiation temperatures <550°C and so are not representative of HTR temperatures

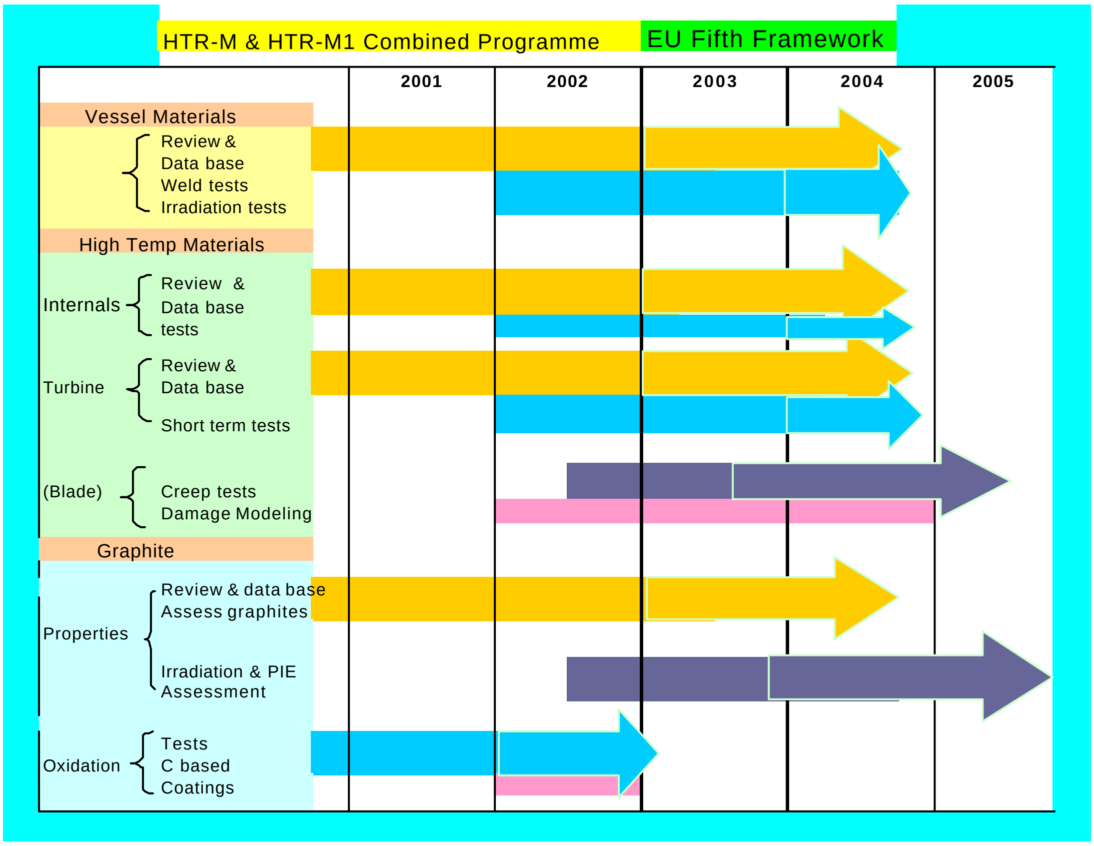


FIG. 1 Description of Overall HTR-M & M1 Materials Programme

3. Results

3.1 Reactor Pressure Vessel (RPV)

3.1.1. Main results from materials review & database work

The technological review considers the materials used for developing reactors and experience from other reactor systems. The RPV forms part of the pressure boundary for the helium coolant during normal and abnormal conditions, provides structural support and alignment for the components within it (core and core support structures) and can operate at temperatures higher than 450°C. For such an application information is required on materials behaviour during manufacture, in the operating environment (neutron-irradiation fluence, operational temperature, and helium environment) and on welding for assurance of integrity.

Two types of materials are currently considered for the HTR RPV:

- SA 508 and its European derivatives, Mn-Ni-Mo or C-Mn type steels: these are used for the 'cold' (cooled) vessel concept similar to that adopted for the PBMR reactor. The advantage of these steels is the large experience that can be taken from LWR vessel technology and design rules.
- Cr-Mo steels with modified 9Cr1Mo steel grades similar to ASME Grade 91 being the prime candidate: These are used for the 'warm' vessel concepts such as adopted for the GT-MHR

reactor which are intended to operate at 400 – 460°C with potential temperature excursions up to 570°C.

These two types of steels have similar strength levels at temperatures up to 370°C but above 450°C allowable stresses for all materials fall off rapidly. Modified 9Cr 1Mo steel allows higher temperatures with only a gradual reduction in design strength at temperatures up to 450°C.

For the HTR-10 test reactor in China, silicon killed C-Mn steel to ASME SA 516-70 is specified which is similar to the C-Mn steels used in the UK for the construction of both steel and concrete pressure vessels in CO_2 cooled reactors of the Magnox and AGR types. For the HTTR in Japan the RPV temperature is close to the core inlet temperature of 400°C and therefore 2¼Cr 1Mo steel has been used for the vessel. Another option is the 12Cr steel adopted in Japan for Fast Reactor steam generator applications and used extensively in steam plants in Europe. For this material welding is more difficult and although some thick section components have been made, most of the service experience relates to thinner sections (pipes, headers). Although 12Cr has a slightly higher strength (10%) below 500°C, modified 9Cr 1Mo steel has superior creep strength at higher temperatures. Modified 9Cr 1Mo steel is considered to have the best potential for both large and small reactors within the belt-line region of the vessel running at the core inlet temperature. Modified 9Cr 1Mo steel is not yet qualified for pressure vessel use according to most design codes. The RCC-MR design rules for steam generator tube material in modified 9Cr 1Mo steel for example were extended during the European Fast Reactor (EFR) project to cover its use for tube plates, shells and hemispherical ends in steam generators with outlet steam temperatures near 500°C but not for vessel applications.

The main safety and structural integrity concerns for the RPV are at the vessel welds, at thicker sections, hot spots, the belt line and regions important to functionality. Fracture, fatigue and creep-fatigue (depending on temperature) are the main damage mechanisms, and degradation mechanisms (irradiation, thermal ageing, temper-embrittlement and corrosion) are the main environmental considerations. The effects of irradiation on the toughness of LWR steels have been studied extensively, the major effect being an increase in the ductile brittle transition temperature (DBTT) due to the combined effects of matrix hardening and grain boundary weakening. This is often associated with the residual elements phosphorus (P) and copper (Cu). For LWR this threat is generally considered to be significant when irradiation causes a change from ductile to brittle fracture at temperatures within the operational envelope. For the warm vessel option, no data are available for modified 9Cr1Mo steel irradiated under conditions expected for the HTR. Information has however been obtained at much higher doses (=1dpa) and these show a strong effect of irradiation temperature in the range 250-450°C. Effects may be small on ?DBTT at 400-440°C but measurable at 350°C. There is no information on the behaviour of modified 9Cr 1Mo steel welds.

On thermal ageing embrittlement (TAE) the base materials and weld metals of both cold and warm HTR vessel steels normally have very fine grain sizes and therefore the most likely embrittlement location is the coarse-grained heat affected zone. This is a narrow region adjacent to the fusion boundary where temperatures of 1100 to 1300°C are reached during welding. Cold vessels operate at temperatures below the embrittlement range (350-550°C) so thermal ageing embrittlement is only possible during transients involving exposure to higher temperatures for tens or hundreds of hours. “Warm” vessels on the other hand operate in the TAE temperature range throughout their lifetime and therefore TAE is of greater concern with regard to vessel integrity. For a material such as modified 9Cr 1Mo steel operating in a “warm” vessel environment, thermal ageing effects in 40 years at 450°C may be significant but useful experiments on this may not be possible within the time scale of the HTR-M project.

For material compatibility, the main concerns are on metal loss/ carburisation/ decarburisation due to actions of the impurities in the helium coolant. HTR gas chemistry is dominated by the impurities H_2, H_2O, CO and CO_2. Low concentrations of H_2O and H_2 are produced by leakage and / or de-sorption from both metal surfaces and graphite. CO, CO_2, and CH_4 may be produced by coolant / graphite reactions at high temperatures. Information on He impurity levels has been assessed from various sources and thermodynamic assessments made with the results of the assessments in terms of the carbon activity and oxygen potential (notional partial pressure of oxygen) introduced on to phase diagrams for Iron and Chromium. For the HTR vessel, metal losses will be either by controlled carburisation in cold wall vessels, and decarburisation combined with internal oxidation in warm wall vessels. The kinetics of these processes will determine the effective metal loss. Overall however, coolant compatibility issues are not expected to have a strong influence on vessel materials selection because metal loss allowances will be a small proportion of the pressure vessel wall thickness.

An important objective of the HTR-M project is the development of a database of materials information for the key components, which includes the RPV. The database is intended to jointly cover vessel steels, high temperature alloys and graphites. Several materials have been considered including those discussed above and aspects such as composition, manufacturing information, test and design data plus environment have been included in the design of datasheets for recording information. The materials property database includes code data where available, and raw data for analysis. Significant amounts of data exist for all these vessel steels and the main focus for this component has been on comparisons and assembling of relevant design data. The design property information will also help drive future decisions on information needs and tests and provide the basis on which the database will be built up and expanded in the future. The Alloys-DB web based system, is currently developed for high alloy steels and is being investigated as a potential means of housing the information on the web in the long term. Such a system allows remote partner access yet maintains a secure transfer of information between the different partners and countries. The system currently includes metallic materials (vessel steels and high temperature alloys), and is to be developed to cover c-based materials (including graphite).

3.1.2. Specific tests on irradiated and non-irradiated RPV welded joints

The above considerations confirmed modified 9Cr 1Mo steel as having the most uncertainties with respect to available database, fabrication experience and HTR vessel characteristics. This material also potentially has the wider application in terms of temperature operation. Modified 9Cr 1Mo steel was therefore selected for the specific experiments on welded joints under irradiated and non-irradiated conditions.

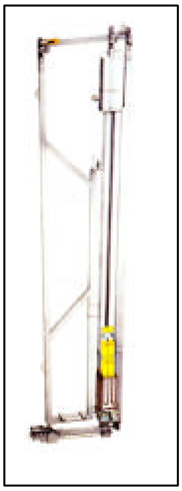
The objective of the irradiation experiment is to achieve the End of Life (EOL) neutron fluence with a relevant neutron spectrum for the HTR pressure vessel at the nominal operating temperature. The EOL neutron fluence depends on the design and type of core. The conditions were estimated as follows:

- Peak neutron fluence – $8x10^{22}$ n/m^2 (E>0.1 MeV)
- Nominal irradiation temperature: 375^{o}C +/- 10^{o}C
- Irradiation to take place in the poolside of HFR (Petten).

The irradiation experiment uses equipment developed from a previous design of Lyra test rig (see Figure 2) used in HFR poolside irradiations, which allows accurate control of temperatures in inert atmospheres. Figure 3 shows details of the loading (specimens). Tungsten plates shield the specimen holder from gamma radiations to avoid high gamma heating. Heaters in the rig are used to maintain nominal temperature conditions.

For the tests, some representative thick section welded plates were manufactured (by Framatome) using narrow gap TIG, a double V weld preparation and a reduced phosphorus filler material. The fabrications mirror the production procedures expected to be used for the HTR RPV welds. The fabrication and characterisation of the welded specimens involved tests on 40 & 150mm thick plates. The thinner plate specimens provide a means for trials using different weld bead lengths and different weld parameters before producing the final weld specimens with the thicker plate. The irradiation conditions chosen for the tests was 10mdpa selected to simulate what was considered to be an upper bound to the expected the EOL accumulated fluence in the belt line region. The test pieces were manufactured from different areas of the welded plates and assembled within the Lyra test facility (see Figure 3).

The test Programme includes tensile, impact, fracture and creep tests in both the irradiated (in shielded facilities) and non-irradiated conditions. Round robin tests were also carried out to check the consistency of testing practices and results between the two sets of facilities used.



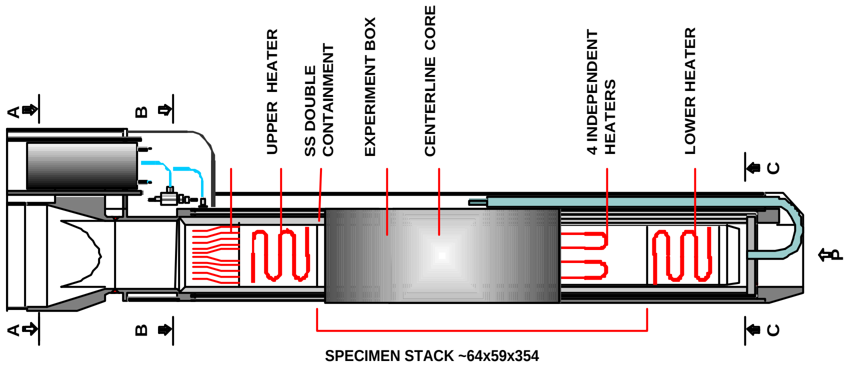


FIG. 2 Lyra test rig used for Irradiation of vessel material within the High Flux Reactor(HFR) at Petten.

The Lyra test facility, (see figure 2) provides accurate control of the temperatures and use of a helium atmosphere. Heaters in the rig correct the temperature levels obtained from gamma heating. The rig was assembled and loaded into the reactor and given that in the poolside region the maximum fluence rate per cycle is 5mdpa, for 10mdpa two cycles loading cycles were used with the test pieces rotated by 180^o between each cycle.

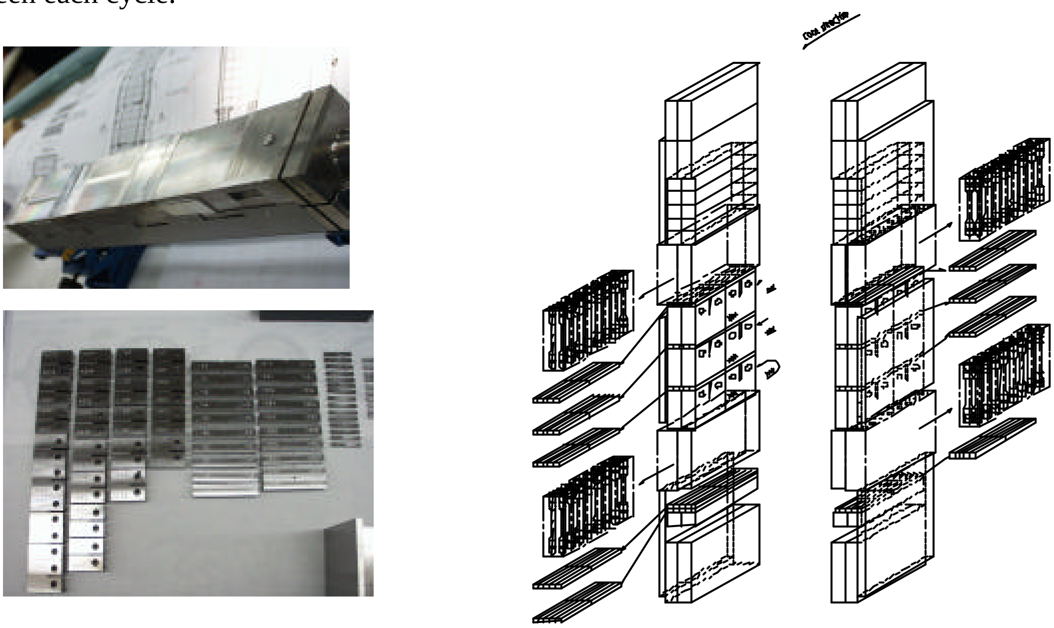


FIG. 3 Test piece assembly within specimen stack of Lyra test rig

The actual temperature of the specimen was monitored using thermocouples at different levels within the irradiation rig. The fluence gradients resulting from the flux buckling within the reactor and self shielding was determined using neutron monitors. The specimen fluence gradients were kept low (<5%) and the notch axis maintained parallel to the HFR core box. Pure helium was used for the environment in the test.

The vessel irradiation finished in March this year and PIE on the specimens started in May 2004. The testing should take around 250 days. PIE consisted of tensile, fracture, toughness and impact plus creep tests within NRG shielded facilities. Most of the test will be finished before the end of the year, except for some creep tests and some of the microscopy. Results from the experiments are currently being assembled and compared. After testing the specimens will be kept at NRG laboratories for another two years before disposal. Not all the irradiated specimens will be tested during the 5FP programme. Some further tests are planned within the 6th Framework Programme on 5FP-irradiated specimens to widen the results for the database.

3.2 High Temperature Materials –turbine disc & blade:

3.2.1. Control Rod

3.2.1.1. Main results from materials review & database work

For non-graphite internal components (e.g. core support plates, grids, etc.) austenitic and some ferritic steels can be used. Such materials have an established capability at temperatures up to 550°C and have been used within the reactor block of other high temperature reactor projects (e.g. Advanced Gas Cooled Reactor in the UK, the European Fast Reactor Project and High Temperature Reactor Projects such as AVR). For higher temperatures however, nickel based and Fe-Cr-Ni Alloys need to be considered.

For the Control Rods, Alloy 800 has been adopted for past and current test reactors (e.g. HTTR and AVR). The control rod compensates for fuel burn-up and power variation reactivity effects and is used to control the reactor operation under fast normal operating modes and to shutdown the reactor system. For very high temperatures (VHTR's) available metallic materials (such as alloy 800H) are considered to be at their operating limit and for such applications, carbon-carbon composite materials offer increased thermal resistance and improved reactivity control during shutdown.

The use of carbon-carbon composite materials for control rods is not a proven technology. The material sits permanently in a neutron field accumulating fast fluences that change its structure and properties. As with other carbon based materials (and graphite), significant changes in the physical and mechanical properties occur due to the effect of irradiation and temperature. A technical synthesis of published information showed that properties are largely an-isotropic and severely degraded by neutron irradiation. Also a specific review of experience was available in Germany from earlier developments for HTR ducting applications.

From the references studied, only limited information on irradiation induced properties appears to be available (mainly from Fusion applications) and although some general trends were seen in properties such as thermal conductivity and thermal resistivity properties differ from one material to another. Also the high cost and limited supply of suitably manufactured material was prohibitive. Given therefore that the selection of the material is very important (and costly) it was decided to involve manufacturers at an early stage (as with graphite manufacturers) and agree with them a suitable material that can be irradiated and tested in a limited way in the HFR. Such tests are planned for the 6FP. The information gathered within this project nevertheless provides a basis for recording available published information and assessing likely needs and property requirements for future tests.

3.2.2 Turbine disc & blade

3.2.2.1 Main results from technical synthesis & database work

The review considered eight potential materials for the turbine disc, thirteen for the blade (including the Russian backup options) and three structural materials for the stator and ducting, etc. Creep resistance and high temperature strength were important requirements and expected to be a main selection criterion

High temperatures (850-900°C) and long-term endurance (60,000h) are key issues. The main concerns are creep and the influence of the environment. Manufacturing considerations are especially important for the disc and blades. Candidate alloys for the disc must have good forging properties and proven thermal stability. For the blades cast material is not sufficient, and so directionally solidified or single crystal alloys have to be considered. The review suggests that the need for cooling is an important issue for both disc and blades. For today's disc materials temperature limits are in the region of 750°C, suggesting that a cooled disc could be necessary for the design.

The impact of corrosion is an important selection criterion since the coolant gas used in a HTR contains small levels of impurities (H_2, H_2O, CO, CO_2) at low partial pressures, and these can interact with the core graphite and metallic components and cause some degradation of their properties. Low concentrations of H_2O and H_2 are produced by leakage and/or desorption from both metal surfaces and graphite. CO, CO_2, and CH_4 may be produced by coolant/graphite reactions at high temperatures. Depending on the partial pressure of the impurities the resulting atmosphere can either be oxidizing or carburising for the selected materials. Damage may be either at the surface or may diffuse to significant depths in the metallic matrices (internal oxidation or carburisation or decarburisation) resulting in a loss of mechanical properties over a significant thickness of the material. The presence of alloying elements such as cobalt (which is in most of the currently available turbine disc and blade materials) is a further issue as is the potential for plate-out and lift-off of particles and their activation and prevention of going through the core.

The review and data base work identified some potential materials to be considered for the experiments. Investigation of turbine disc materials revealed Udimet 720 to be the best currently available candidate that would not require significant manufacturing development. For the perceived endurance this is thought to have a temperature ceiling of around 700-750°C and is likely to require cooling. Two DS grades of material were selected for the blade tests (CM 247 LC as an Al-oxide former and IN 792 as a Cr-oxide former). Because of corrosion considerations, the current view is that any blades may have to be coated for long life. Application of suitable coatings can arrest creep reduction tendencies, but their use requires an understanding of the potential for interfacial cracking at the coating layer to avoid the development of more significant cracking from the interface. Coatings must however be of the same type (chromium oxide former or aluminium oxide former) as the base alloy. The choice of coating remains to be made and tested in a future Framework Programme. Adoption of a cobalt free material was not considered a priority following investigations made by HTR-E [3].

3.2.2.2. Specific tests on selected materials

The planned experiments on these turbine materials involve short and intermediate high temperature mechanical/ creep tests in simulated environments. The short-term tests on the disc material were carried out at 700 and 750°C. For the turbine blade materials short and intermediate tests were carried out at temperatures up to 850°C. The duration of the intermediate creep tests is around 3000h and performed on one of the turbine material grades and will also include aged and notched samples.

Procurement of suitable quantities of the materials have been made and characterization tests on the disc and blade materials completed or underway. The test matrix and proposals for the test conditions as far as possible bound the temperature and transient cases expected by the turbine. Four environments are used to pre-treat the materials (see Figure 4) before testing in air and/or argon.

- As received with ageing heat treatment – I
- Decarburised – II
- Carburised – III
- After the carburisation or decarburisation heat treatment – I(mod)

Currently high temperature mechanical/ creep tests have been completed in air for the disc material and for blade material CM 247 LC DS. Further tests are planned after the conditioning of the remaining specimens is complete. To-date the results suggest that strength values (tensile and creep) are consistent with published information. Fatigue results (for the disc material) are dependent on grain size but nevertheless better than expected. The start of the medium term creep tests on the treated specimens will begin as soon as practical towards the end of 2004.

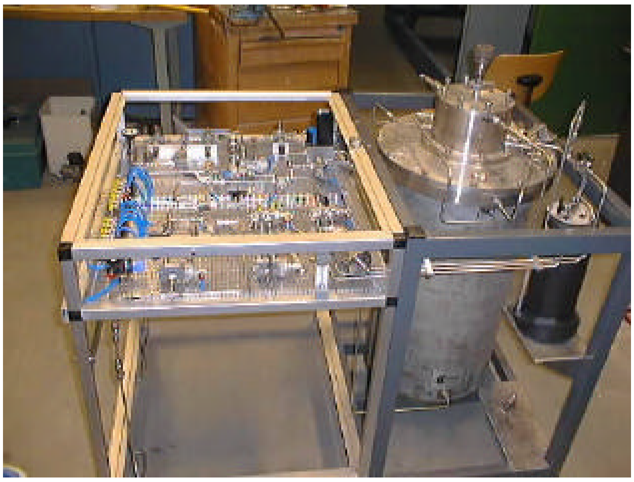


Figure 4 Carburising test rig

3.3 Graphite core:

3.3.1 Main results from materials review & database work

The graphite core is a key component that affects safety and operability of the reactor. It provides structural support, coolant channels, moderation, and shielding while operating in a high temperature helium environment. Its performance is critically dependent on the graphite properties, which are irradiation dependent. The final choice of graphite for the core should be based on a number of factors, although the most important will be the effects of fast neutron irradiation on it's properties up to the peak doses envisaged. Given the best graphite available, it is the task of the core designer to produce a design that will operate safely over the design life of the reactor. The most important considerations are component integrity and changes in core geometry, both of which are affected by the dimensional change (see Figure 5).

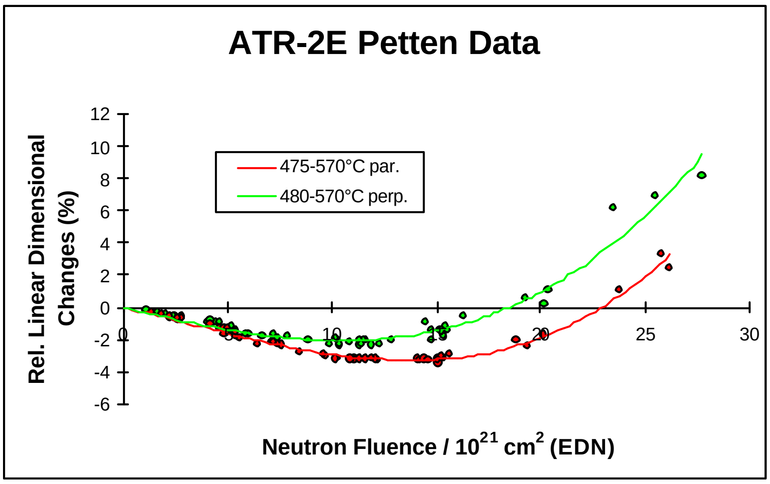


Figure5 Effect of irradiation on dimensional change

Many of the graphites used in previous core designs are no longer available and there has been a serious decline in ability to manufacture nuclear grade graphite in large quantities. The main questions concern the long term availability of the coke and manufacturing procedure. Current HTGR projects - HTTR (Japan) and HTR-10 (China) - use a Japanese graphite called IG-110, which, with its high strength, is suitable for exchangeable core components where low fast neutron fluences are applicable.

Much of the available irradiated materials data are obtained from various materials test reactors and often it is not possible to complete an irradiated materials property database for a particular graphite in advance of design and construction phases of a reactor. In such cases design databases can be used to arrive at the required information, developed from previously irradiated graphites of similar microstructure, and an understanding of irradiation damage in the polycrystalline graphites. R & D activities have been undertaken in a number of countries to investigate graphite properties. The INGSM International Nuclear Graphite Specialist Meetings provide a valuable forum for the exchange of information and opinion on graphites and has recently held seminars to review the current situation.

Two of the most significant experimental programmes in the past were for the European Dragon HTR programme, and the German HTR programme. Unfortunately most of the data obtained by other countries are confidential and so far are not published in open literature. The work on the review and collection of graphite properties considers the accessible information on the IAEA database, internal information and published information at seminars and conferences. The IAEA database was established to help the development of International programmes on graphite-moderated reactors, assist safety authorities in assessment of safety aspects and serve as a source of scientific information for nuclear and non-nuclear technology.

The data relevant to the irradiation temperature and neutron fluence domains for new HTR's will largely come from the new tests and be built up for each graphite grade and sample orientation, and contain the appropriate details of the graphite i.e. grade, manufacturer, coke source, grain size and manufacturing method. These data are being collated onto a set format for use within the project. It is planned to present the experimental data on the new graphites in the form of recommended 'design curves' for each required material property.

For nuclear applications, the graphite has to be as free as possible from impurities. Most of the impurities present will become activated during the operating life of the reactor, which will give rise to operational problems, as well as decommissioning and final disposal problems. Most impurities, however, are volatile and so disappear during graphitisation. To remove as much of the remainder as possible, halogens are added generally during graphitisation to aid the conversion of metal impurities and boron particularly, to their more volatile halides. (Extremely low boron levels are important from a reactor physics point of view, as it is a very strong neutron absorber.)

Meetings and discussions have been held with graphite manufactures and there has been much interest in providing materials for the test programme. Suitable graphites were made available by Graftech and SGL who have offered extruded and iso-moulded graphites for the irradiation experiment. There has also been interest from other graphite manufacturers to supply material for the experiment and JAERI have offered international collaboration through Toyo Tanso who have proposed to supply the graphites they use in the HTTR reactor. It is clear that further input to graphite selection will be strongly influenced by the outcome of the planned irradiation experiment discussed below.

Work carried out in Germany to develop SiC coatings for graphite materials for protecting the graphite against oxidation from air or steam ingress under severe accidents has also been reviewed. Investigations were done using the matrix German graphite A3-3 to measure corrosion rate for temperatures up to 1200°C, and on coated pebbles using different graphites. Most of the coating experiments used the Japanese graphite IG 110 adopted for the structures and fuel blocks of the HTTR. Corrosion tests, irradiation tests plus by PIE were carried out to assess coating integrity and different coating methods (chemical vapour deposition (CVD), paste siliconisation (PS)). A very good corrosion resistant quality was eventually achieved with IG 110 and V438 using CVD. No damage or weight loss was observed after being irradiated and corroded and dropped from a height of 50 cm. Although the original objective, to develop a coating for A3-3 graphite, was not achieved, these results gave a very positive indicator and provide a useful basis for further investigation.

3.3.2 Specific tests on irradiated and non-irradiated graphites

3.3.2.1 Oxidation tests

This experimental work uses the induction furnace facility INDEX and the thermo-gravimetric facility THERA (see Fig.6). Graphite burning under severe air ingress accidents, is usually assessed using computer models, based on isothermally measured kinetic equations that consider in-pore diffusion and chemical reaction mechanisms. The data requirements for such models are burn off dependent chemical reactivities (regime 1) that occur at lower temperatures and consumption of oxidising gas within the pores (regime II) that becomes more important at intermediate temperatures and the oxidation attack becomes smaller in the depth of the material. At high temperatures external mass transfer is important and the reaction is restricted to the geometrical surface only. THERA was used for the regime 1 and INDEX for regime II where higher flow rates are needed. Long-term experiments at low temperature in air were also performed in an annealing furnace.

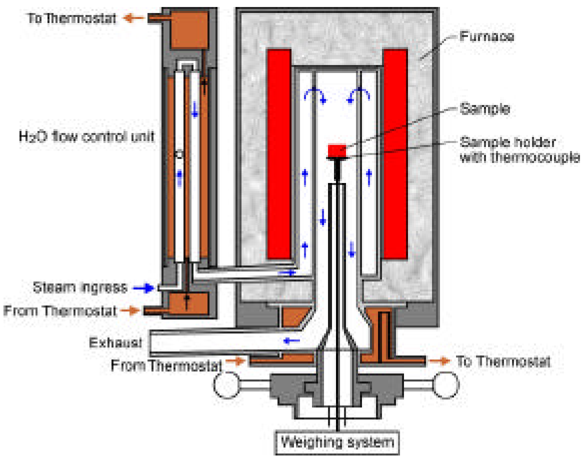


Figure 6 Schematic of THERA test facility

The second series of experiments looked at the performance of CFC materials under different temperatures in steam and air. Oxidising gas flowed through the inner bore hole of the tube with flow rates sufficiently high to suppress the influence of boundary layer mass transfer on kinetics. Oxidation effects were measured by mass spectrometric analysis of the product gases and by weight loss of the sample. Experiments were also performed to determine properties of CFC materials irradiated (up to 1dpa) and non-irradiated. These include measurements of strength, dimensional change, thermal diffusivity and heat capacity. The main findings from this graphite oxidation work are summarised as follows:

- The un-graphitised binder material has to be treated as a separate component because of its remarkably different oxidation behaviour.
- Despite its Si-content, the oxidation resistance of NS31 is not notably better than that of other examined materials (but a factor of 2 better than the case without Si).
- Both the amount of Si and its distribution within the material determines its ability to protect against oxidation
- The oxidation behaviour of homogeneous materials in regime 1 is characterised by a continuous rate verses burn-off curve with one (more or less) pronounced maximum.

- The shape of the rate vs. burn-off curves is roughly similar in air and steam, however the rates differ markedly. For regime 1: for a similar rate in air at 600 – 650 °C, temperatures are required to be 250°C higher in steam (900°C)
- Inhibition by hydrogen/ CO remains to be measured for steam oxidation.

3.3.2.2. *Irradiation tests*

For these tests three main graphite manufacturers were approached to see which graphites could be offered for the next generations of reactors. Five main grades of graphite as highlighted in the table, were selected and identified for the programme. The major graphites chosen were NBG-10 and NBG-25 from SGL and PCEA and PCIB-SFG from Graftech. A range of grain sizes (1mm down to 10 µm) was selected. The IG110 (HTTR) graphite was also chosen (iso-moulded graphite made using a petroleum coke) since this has previously been irradiated at high temperature. Although for use only to a small/medium fluence it would still provide a useful reference material for the experiment. In addition to these five, other potential 'minor' graphites were identified for inclusion as 'piggy-back' samples in the irradiation capsule (Table 1) in order to provide an early indication of their irradiation behaviour. The (minor) graphites selected were Graftech grade PPEA, SGL grade NBG-20 and Toyo Tanso grade IG-430. It has also been possible to introduce extra materials into the experiment for microstructure evaluation and X-ray/ tomography.

SGL	PITCH COKE	PETROLEUM COKE
EXTRUDED	**NBG-10**	NBG-20
ISOMOULDED		**NBG-25**
GRAFTECH	PITCH COKE	PETROLEUM COKE
EXTRUDED	PPEA	**PCEA**
ISOMOULDED		**PCIB-SFG**
TOYO TANSO	PITCH COKE	PETROLEUM COKE
EXTRUDED		
ISOMOULDED	IG-430	**IG-110**

Table 1 Graphites selected for the irradiation test programme

The irradiation of the graphite samples is carried out using the INNOGRAPH-1 test rig (Figure 6) inserted in the core of the HFR. For a HTR, the temperature range of interest is 550-950oC. The peak fluence will be at around the mid-range temperature of 750°C and so this was chosen for the first experiment. It was clear that exposure up to full fluence would be needed but this would not be possible within the programme timeframe. It was necessary therefore to plan the tests so that the irradiation could be carried out in two steps, the first over the 5FP (HTR-M1 programme) and the second within the 6FP (V/HTR-IP programme) (see Figure 7). This means that the experiment was designed to enable the samples to be withdrawn and tested and then re-inserted in a new rig for continuation of irradiation exposure within HFR.

The target flux in the current four-year programme is as high as possible. The current available locations in the HFR suggest an irradiation for 1-1.5 year to an EDN fluence ~6-7 10^{25} m-2 (approx. 8 dpa)). This is not expected to yield properties up to turn round behaviour but will provide information on whether the graphite is useful or not. The current framework therefore provides a first sorting and as the experiment progresses there is the possibility of replacing those that are unsuitable with alternatives.

The irradiation test provides data over a range of fluences within a single test run by using the flux (buckling) distribution available in the HFR capsule. A third order polynomial can describe the behaviour and a sufficient number of points should be obtained from the test to describe the variations and uncertainties in the properties. The temperature is controlled by a combination of He/Ne in gas gaps and gamma heating and the rig was designed to give a homogeneous temperature profile in the specimens.

	Planned			**Realised**		
	WG	AG	Total	WG	AG	Total
NBG-10	15	15	30	**15**	**15**	**30**
NBG-25	15	15	30	**15**	**15**	**30**
PCEA	15	15	30	**15**	**15**	**30**
PCIB	15	15	30	**15**	**15**	**30**
IG-110	15	15	30	**15**	**15**	**30**
NGB-20	5	5	10	**7**	**7**	**14**
PPEA	5	5	10	**7**	**7**	**14**
IG-430	5	5	10	**9**	**9**	**18**

Table 2 Comparison of planned and realised graphites in INNOGRAPH-1

The rig design requires the stacking of up to 150 samples, 8mm diameter (either 6 or 12mm in height), covering the two main directions parallel and perpendicular with grain (15 samples per grade per direction). Figure 7 shows the assembly details of the drum-based configuration. The specimens were in four stacks with 60mm stacking height per drum and separated from surrounding TZM alloy with graphite foils. Characterisation tests were carried out prior to loading in the test rig to determine properties before irradiation. These cover dimensional change, CTE (coefficient of thermal expansion), dynamic Young's modulus and thermal diffusivity. For CTE a commercial dilatometer was used. Following completion of the characterisation tests the samples were loaded into INNOGRAPH-1 and it was possible through careful arranging of the specimens to realise a greater number of piggy back specimens to those first envisaged (Table 2).

Figure 7 Assembly of INNOGRAPH-1 – details of drum based configuration

The Irradiation started in February 2004 and is currently in progress. The first PIE will be in the spring/summer of 2005. To-date the experiment is continuing satisfactorily although a slight non-uniformity of temperature in the two lower drums has resulted in a reduction in nominal temperature (670°C) for the samples in these drums. Because the sample temperatures can be individually monitored results at 750°C(75%) and 670°C(25%) will be available. Over the coming months decisions on what specimens to irradiate during the second phase will be made.

4. Conclusions & further work

The objective of the HTR-M & M1 projects is to improve knowledge of materials for future HTR's for the reactor pressure vessel, the control rod and turbine (blades & discs) and the reactor graphite. The scientific and technical progress for the HTR-M project has been described and issues that are likely to influence the technical decisions on the next stage of the programme reviewed. The review and data base work is largely completed and testing is in progress with the main test on the vessel and graphite irradiations in progress.

The vessel review work has focused on materials used in new and developing plants and identified Modified 9 Cr 1 Mo steel as a material with important potential for the VHTR. A key requirement is to confirm its suitability for RPV application taking account of the environment. This material was recommended for irradiation tests. The pre- and post irradiation test programmes are largely complete with PIE testing in its final phase. A comparison of results should be possible over the next few months. Future work in FP6 will then be to consolidate this database through further tests on pre-irradiated specimens at different creep temperatures and to perform larger scale non irradiated creep tests under representative vessel loadings and geometries

The review and database work on high temperature materials relevant to the control rod and turbine disc and blade materials are largely complete. Carbon-carbon composite materials are recognised as a higher temperature alternative to metals for the control rod. For the turbine (disc and blades) candidate alloys have been selected for the test work. Testing has been completed in air showing a good agreement with manufacturers properties. Pre-treatment of the specimens to determine the effect of carburisation and de-carburisation is in progress and testing of the treated specimens should be completed before 2005. Intermediate creep testing of the material is planned to start shortly under the HRT-M1 project. Further work on high temperature materials in FP6 will focus on the material selection for the heat exchanger and the completion of short term qualification tests to confirm selected materials.

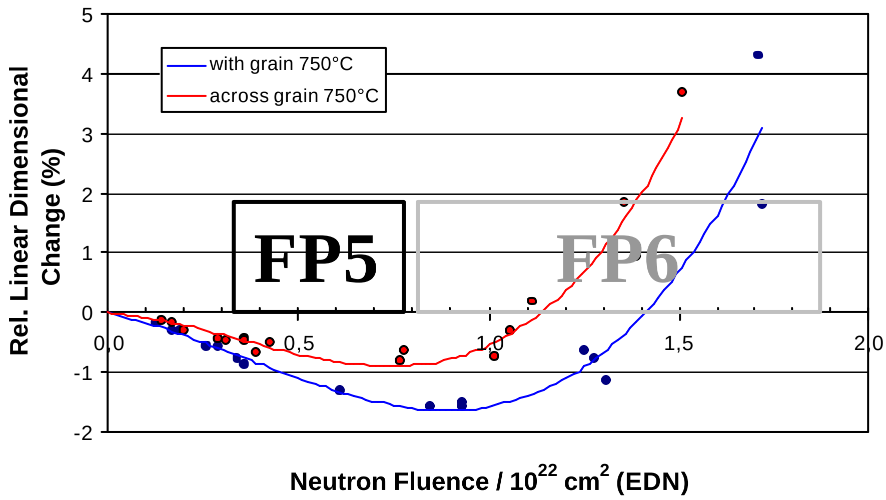


Figure 8 FP6 builds on the FP5 graphite results

The work on the graphite property side continues following a review of available information on former graphites and oxidation tests on graphite and c-c composite materials carried out at FZJ using hot air and steam. Three principle graphite manufacturers and suppliers have collaborated closely with the HTR-M1 project and supplied new graphites for irradiation testing in HFR. Following selection, procurement, machining and pre-characterisation of the graphite samples, and assembly of the INNOGRAPH-1 test rig the irradiation experiment is now underway and the first phase (FP5) expected to be completed in 2005. A second phase (FP6) will begin as soon as possible afterwards with the view of reaching the full fluence condition and defining the property curves beyond turn-round (Figure 8). It is also intended to carry out a further test at a higher temperature (>950°C) during 6FP to full fluence and support this with further oxidation tests and micro-structural modelling investigations. The latter is considered necessary in the longer term to reduce the need for extensive testing of new graphites and to provide the basis for a specification for graphite development. Also planned is the development of design guidelines for graphites and c-c composites, which can be used to support design activities on future graphite cores.

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