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Investigation of High Temperature Reactor (HTR) Materials

by

D.E. Buckthorpe	NNC Ltd.	, UK
R. Couturier	CEA	, France
B. van der Schaaf	NRG	, Netherlands
B. Riou	FRA	, France
H. Rantala	JRC	, Netherlands
R. Moormann	FZJ	, Germany
F. Alonso	EA	, Spain
B-C. Friedrich	FRA	, Germany

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D. Buckthorpe
NNC Ltd., UK

R. Couturier
Commissariat à l' Energie Atomique (CEA.), France

B. van der Schaaf
Nuclear Research and Consultancy Group (NRG), Netherlands

B. Riou
Framatome ANP, France

H. Rantala
European Commission, Joint Research Centre (JRC), Institute for Advanced Materials, Netherlands

R.Moormann
Forschungszentrum Juelich GmbH (FZJ.ISR), Germany

F. Alonso
Empresarios Agrupados Internacional S A (EASA.MD), Spain

B-C. Friedrich
Framatome ANP (GmbH), Germany

Abstract

Issues associated with material selection and behaviour are being examined for the main reactor circuit components of the Modular High Temperature Reactor. The work is being performed as part of a European development supported by the European Union Fifth Framework programme. The work considers materials for the reactor vessel and certain high temperature regions including the turbine and the graphite core. This paper reviews the main elements of the materials work programme and reviews some initial findings with regard to material choices and needs.

Introduction

A common European approach to the renewal of HTR technology has been established through the direction of a European HTR Technology Network (HTR-TN) to coordinate and encourage work-shared structures and serve as a channel for international collaboration in this technology [1].

The European Commission is supporting a number of research activities on modular HTR technology in its 5th Euratom Framework Programme[2]. These involve partnerships between principal industrial and research organisations from countries of the European Union working to bring European expertise and experience in the HTR together and to support industry in the design of reactors. Two Projects within this Framework programme HTR-M and HTR-M1 consider the selection and development of materials for the key components of the HTR. They extend over four years and involve eight partners. This paper gives an overall description of the HTR materials projects, their objectives and some results from initial actions that focus on the needs for feasibility with respect to material choice and structural integrity.

Background to HTR development in Europe

Commercial experience with Gas-Cooled Reactors began in 1956 in the UK which extended to cover 26 Magnox Stations and 14 Advanced Gas-Cooled Reactors using CO_2 gas as a coolant. HTR plants began in Europe in the 1950s. These used helium as the coolant to permit an increase in operating temperature. Coated ceramic fuel particles and a dispersed graphite matrix and moderator were used and in the years 1960 to 1990 many HTR prototypes were built and tested in Europe (DRAGON (UK) - 1964 to 1975, AVR (Germany) 1966 to 1988, THTR 300 (Germany) 1983 to 1989). These provided a valuable foundation on which to base future HTR development.

There was a continued interest in larger steam cycle plants in Germany, Russia and the United States through the 1970s and commercialisation seemed promising in the early 1980s with the medium sized concept developed in Germany for industrial process heat applications. The commercial development of this HTR-MODUL plant was however terminated in 1990.

Renewed interest in HTRs in Europe came following the development of the HTR coupled to a gas turbine power conversion system which led to todays developments of the industrial prototypes GT-MHR (in Russia) and PBMR (in South Africa) which are supported by International teams with a strong European involvement.

Overall Objectives and description of the HTR Materials Projects

The main objectives of the HTR materials projects are to provide information for the key components and support the development of High Temperature Reactor technology in Europe. Two projects are ongoing (HTR-M & HTR-M1 -managed together) that cover three areas: the Reactor vessel, High Temperature Materials (internal structures and turbine), and Graphite Structures including oxidation. A description of the main activities and tests to be performed is given in fig. 1.

Reactor Vessel

Design information for the HTR Reactor Pressure Vessel (RPV) is not well established under HTR conditions and further improvement is needed in the areas of design and structural integrity analysis and materials properties data. Also information is needed on manufacturing of products and parts, behaviour of welded joints and the influence of environment (neutron-irradiation fluence, operational temperature, and helium environment). The integrity of the welded joints is seen as a particularly important feasibility issue. The objective of this work therefore is to investigate and confirm the choice of candidate material options for different HTR concepts, in relation to both normal and accident conditions, and to compile their design properties.

[1]. W. Von Lensa , D. Hittner, J. Guidez, F. Sevini, A. Chevalier, M. T. Dominguez, G. Brinkmann, T. Abram, J. Martin-Bermejo. "The Role of International Collaboration within the European High-Temperature Reactor Technology Network (HTR-TN)" Seminar on HTGR Application and Development, Beijing, China, March 19-21, 2001.

[2] J Martin-Bermejo, M. Hugon, G. Van Goethem
Research activites on High Temperature Gas-cooled Reactors (HTRs) in the 5th EURATOM RTD Framework Programme. SMiRT 16, Washington, Aug 2001

The work (performed in HTR-M) covers a review of vessel materials and their properties, focusing on existing thermal gas-cooled reactors and previous high temperature reactors and the setting up of a materials database on design properties. This will lead to a recommendation for appropriate vessel materials, depending on operating conditions, and identification of data omissions and the selection of a material for testing under irradiated and non-irradiated conditions. The Work Package will consider materials for a “cold” (current PBMR) and “warm” (GT-MHR) vessel option. Specific tests on welded joints are to be performed covering tensile, creep and / or compact tension fracture specimens to determine representative properties including the effects of irradiation. A further review and synthesis is planned on completion of the irradiation test work and examination.

High Temperature Materials

The HTR primary circuit components operate at temperature above 600°C and up to 850 to 900°C in order to achieve high energy levels and high cycle efficiency. For such components a number of metallic and ceramic materials are required in the region of the core and reactor internals (i.e. metallic, ceramic and composite materials) and for the core support and other structures. For these materials and components, some data exists but the information and experience are not well established for the HTR environment. Also metallic materials are required for the turbine components for which the expected working conditions are outside todays industrial experience and a significant effort of material development is required in this area.

The first step involves putting together a database of properties that takes account of the experience on existing high temperature and gas-cooled reactors. This is covered in HTR-M. The database will include a reference to material composition, microstructure, manufacture (products and parts, etc.) and extend to materials required to withstand temperature levels experienced in emergency modes (e.g. Carbon based materials reinforced by carbon fibers, Ni-based alloys, etc.). The control rod cladding is investigated for the internal reactor structures and the disc and blades of the turbine. For the turbine, the two most promising grades will be investigated taking account of different options, manufacturing aspects and characterisation of improved alloys made by various manufacturing routes. Short term mechanical tests needed to quantify the materials in the HTR environment will be performed covering tensile, fatigue, short term creep and creep / fatigue tests planned (in air, vacuum, helium) at temperatures up to 1000^{o}C. The results of the test programme will be stored in the database.

Within HTR-M1 the work on the blade materials has been extended to cover intermediate term creep effects to confirm the suitability of the chosen alloys for longer-term high temperature exposure. Testing is planned at temperatures of 850^{o}C with testing times up to 10,000 hrs. The expected number of tests will cover specific grades, fabrication routes, and heat treatments. Tests will also be carried out on aged material and damage analysis and lifetime modelling will be performed (based on results from notched specimens) and the results incorporated into the developing database.

Graphite

The need for reliable graphite data is a crucial issue for existing and new HTR projects and is an important consideration for future decommissioning activities. Graphite plays an important role as a moderator and structural component and has important safety implications because of structural and other property changes that occur when it is irradiated. Its selection also has important consequences for future decommissioning activities. Oxidation resistance of graphites at high temperature also requires special attention due to its relevance for safety analyses of air and water ingress accidents, licensing procedure and normal operation.

The HTR-M project provides for a limited review of existing graphite information. This is to identify gaps in data and testing requirements. Many of the graphites used in previous core designs are no longer manufactured commercially, and currently available grades of graphite are limited to a few specific sources of raw materials. Very few of the grades may turn out to be suitable or desirable for a European based modular HTR. The Work Package also involves test work to investigate the influence of oxidation arising from severe air ingress with core burning and an investigation of protective coatings and innovative C- based materials. Overall this Work Package is expected to provide a platform of material data for future application and selection of graphites for the HTR.

The HTR-M1 project will focus on initiating an irradiation programme on chosen graphites to determine the variations in their physical and mechanical properties up to low/medium irradiation doses (making use of the results from HTR-M as a basis for estimation and extrapolation to higher doses). Discussions will be held with graphite manufacturers to investigate the possibility of producing small quantities of alternative graphites (in the laboratory) which would have a better combination of desirable properties. Whilst this project falls short of the ultimate selection criteria for the graphite, which will mainly be based on long-term irradiation behaviour it will establish the groundwork for continuation of the irradiation programme on the most desirable graphite(s). The project will also make use of valuable diminishing experience from experts in design, experiments and testing of graphite within Europe serving to reinstall graphite irradiation and qualification methods within Europe while the available experience and facilities exists.

Material & Structural Integrity Issues

HTR Reactor Pressure Vessel (RPV)

The work on the RPV will establish available information on a few selected vessel steels and a database for design and feasibility investigations. Two types of materials are currently used for RPVs, depending on whether the temperature of the pressure boundary under normal operation or design conditions exceeds the limit for LWR pressure vessels of 370°C.

Most designs of future plant make use of the either SA 508 steel for insulated vessels or modified 9Cr 1Mo steel for ‘inlet temperature vessels’. These materials have similar strength levels at temperatures up to 370°C. The use of modified 9Cr 1Mo steel allows higher temperatures with only a gradual reduction in design strength at temperatures up to 450°C. Above 450°C allowable stresses for all materials fall off rapidly. Modified 9Cr 1Mo steel has an even bigger advantage over the other materials at these higher temperatures. At 500°C for example, RCC-MR indicates that its design strength is twice that of 2¼Cr 1Mo steel (Fig. 2).

Existing LWR pressure vessels in SA 508 and its European derivatives encompass the conditions in proposed HTR vessels with respect to operating temperature, wall thickness, design features (thick flanges etc.) and irradiation levels. The large database and the extensive fabrication experience from LWR programmes provide a sound basis for HTR vessel design. The most obvious gap in data relates to possible transients involving temperatures above the usual LWR limit of approximately 370°C.

A substantial database and fabrication experience exists at least for CO_2 cooled reactors in the UK, with respect to C-Mn steels similar to the SA 516-70 grade specified in Chinese designs. There is European experience of complementary high temperature materials (2¼Cr 1Mo grades). There should be relatively few problems in assembling an adequate database extending to temperatures involved during transients.

The above considerations leave modified 9Cr 1Mo steel with the most uncertainties as to the available data base and fabrication experience with respect to HTR vessel characteristics. This material also potentially has the wider applicability and since for the Framework Programme it is only possible to test one material within the High Flux Reactor this represents the most likely candidate for the experiments. The extent of testing of control specimens is expected to cover as received (welded and post weld heat treated) and irradiated material (irradiation time approximately 100 - 200 hours). Thermal ageing effects in 40 years at 450°C may be significant but useful experiments may not be possible within the time scale of HTR-M.

The main concerns with regard to structural integrity are expected to be at the welds. They carry an additional requirement to satisfy the safety analysis methodology and safety studies (defect location and defect size) and have to be assessed from the point of view of non-destructive examination and potential for failure or leakage.

The main damage mechanisms to be addressed are fracture, fatigue and creep-fatigue. These have to be evaluated from an initiation aspect and for crack growth of defects under fluctuating thermal and mechanical loads. The potential effects of environment (temperature, irradiation, ageing) all have to be taken into account. The test programme and its development addresses toughness and creep properties under as-fabricated and simulated end-of-life conditions. This will provide an understanding of the welding processes, weld metals and post-weld heat treatment required for thick welds and weld factors to be applied on the base material properties.

Other sensitive zones that are to be checked are areas that feature thicker sections (potential for reduced properties and strength), hot spots and regions important from a functionality point of view. This includes the flange area and belt line which have to satisfy primary and secondary stress limit requirements and progressive deformation mechanisms.

HTR High Temperature Materials

For internal structures austenitic and ferritic steels can be used for some of the non-graphite components (core support plates, grids) where there is an established experience with the steel in gas and other elevated temperature reactors. In general, operability and applicability up to temperatures of 550^{o}C has been firmly established for some materials within projects such as the Advanced Gas-Cooled Reactor in the UK, the European Fast Reactor Project and High Temperature Reactor Projects such as AVR. Some carbon-carbon (C/C) composites have also been investigated for use on HTTR for the control rod. The use of such materials is considered to provide increased thermal resistance offering potential for improved reactivity control during shutdown and for allowing the normal operating temperatures of future reactors to be increased. Experience of materials for operation at much higher temperatures at the moment rely heavily on Nickel based and Fe-Cr-Ni alloys. For the HTR which has the additional effect of a helium environment the PNP and Japanese He/He exchangers provide some important experience (Figure 3).

For the turbine components, especially the turbine discs and blades, criteria for material selection are to be based on a safe operation period, of say 60,000 h at 3000 r.p.m. (50 Hz), with upper temperature limits in the range 850 to 950^{o}C. Candidate alloys for the disc must have a sound industrial base and be capable of production of large defect free ingots with good forging properties and proven thermal stability. Candidate alloys for the disc include A286 (Cr Ni Fe), IN 706, IN718 and UDIMET 720. The introduction of HIP material currently being investigated for the Fusion Reactor also offers the possibility of producing near finished sized components suitable for turbine discs. For the turbine, the materials for cooled and non-cooled blades have to be investigated (although the former clearly offers cost advantages). Metallurgical factors such as cobalt content of the material also have to be assessed with regard to activation potential which may impact on maintenance costs.

The main structural integrity issues concern creep and the influence of the environment. Two basic requirements dictate the design of gas turbine blades and discs, the cycle temperature and the maintenance of critical dimensions (clearances) throughout the service life. The selection of turbine blade designs are often limited by the high temperature creep properties of the material (i.e. non-cooled blade).

For turbine discs there is a requirement to limit the permanent growth and distortion to within typical design life targets. This is usually achieved by maintaining a greater part of the disc cross section within elastic limits, (i.e satisfying progressive deformation or shakedown criteria). Critical regions for the disc from the point of view of potential creep and fatigue failure are the hottest parts. These are at the disc neck because of unrelieved high localised stresses giving rise to undetected growth, and at the rim, due to the additional geometric effects of the blade root slots and possibly cooling holes. Careful material selection or control of localised stress and temperature conditions can avoid creep-fatigue interaction problems in these areas.

For the turbine blades failure modes such as creep, high cycle fatigue and low cycle fatigue have to be taken into account. The gas radial temperature variation often peaks typically in the middle third of the blade with steep temperature gradients across the blade section which vary according to the transient and steady state conditions. All regions of the blade profile therefore undergo complex stress-strain cycling with reversed plasticity in some areas. For material selection a close understanding of some important parameters such as creep rupture, creep ductility and creep rate are needed. Typically for nickel based super-alloys time dependent effects induced by creep and oxidation on cyclic crack growth rates are important for design, and hence an understanding of creep crack growth behaviour and the interaction between creep and fatigue with tensile and compressive dwells is needed. The awareness of the role of grain boundaries in high temperature fracture has also led to significant developments in single crystal super-alloys that can give beneficial anisotropic material properties.

Corrosion can cause a significant shortening of the material creep life and acceleration of creep crack growth rates. Application of suitable coatings can arrest creep reduction tendencies, however 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.

Graphite

a) Properties

The graphite core is a key component that affects safety and operability of the reactor. As well as acting as a neutron moderator and shield, it is a structure that generally provides channels for the passage of fuel and control devices, and coolant flow. The problem is that when irradiated by fast neutrons, graphite suffers damage to the crystalline structure. The resulting damage causes a change in all the physical and mechanical properties of the graphite e.g. strength, coefficient of thermal expansion (CTE), and thermal conductivity.

Ideally, the graphite chosen for HTR designs should be reasonably isotropic, exhibit small dimensional change behaviour, have a low CTE, a high thermal conductivity, a high irradiation creep constant, a low Young’s modulus and high strength. The problem is that many of these properties are not compatible in practice for normal commercial graphites e.g. graphites with high strength also have a high Young’s Modulus. Other factors affecting the choice of graphite would be impurity levels, cost and machinability.

Over the past 60 years, R & D activities have been undertaken in a number of countries to investigate graphite properties. The IAEA International Irradiated Graphite Database Technical Steering Committee is currently compiling a database on graphites past and present and has recently held a seminar [3] to review the current situation, and is currently compiling a database on graphites past and present.

[3]. Graphite Specialists Meeting, Oak Ridge National Laboratory, Oak Ridge, TN, USA, September 5-6, 2000

Many of the graphites used in previous core designs are no longer available in practical terms for a number of reasons. The decline in the ability to manufacture nuclear grade graphite in large quantities in the UK is an example of the above. Two large plants, one owned by Anglo Great Lakes (AGL now SGL) and the other by British Acheson Electrodes Ltd (BAEL, later known as Union Carbide and presently called UCAR) manufactured all the Gilsocarbon graphite used in the UKs AGRs. Neither plant now exists. Todays HTGR projects - HTTR (Japan) and HTR-10 (China) - use a Japanese graphite (IG-110). This graphite, with its high strength, should be taken into account for exchangeable core components of the HTR where low fast neutron fluences and hence low total doses apply. The behaviour for very high doses is not yet fully known.

For a future design it might not be possible to generate a complete irradiated material property database for a particular graphite in advance of the design and construction phases of a reactor. In such cases, existing experience in graphite development together with irradiation test programmes can be used to provide an initial assessment of the suitability of currently available graphites. All available data should be put into a systematic database to find the best candidate materials. These would then have to be irradiated at the appropriate temperatures expected under HTGR conditions and to the peak doses envisaged and screening tests performed, to compare the results with data from the database, and to update the existing models for reliable interpretation and comparison.

Generally, the most important material property change from the point of view of core lifetime integrity is dimensional change. Graphite initially shrinks but then undergoes shrinkage reversal, i.e. growth. This is referred to as ‘turnaround’. The exact form of the dimensional change curve varies with irradiation temperatures. Normally, the higher the temperature the lower the peak shrinkage and the lower the dose at which turnaround occurs. Different graphites can have significantly different dimensional change curves at the same temperatures.

The validation of the through life performance of the graphite core structure is critically dependent on the knowledge of graphite properties and the way they vary under fast neutron irradiation. It is essential therefore that its behaviour is fully understood and investigated up to at least the peak design dose and over the appropriate temperature ranges

The final choice of graphite for the core should therefore be based on a number of factors, although the most important will be the effects of fast neutron irradiation on its 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. For example, graphite shrinkage could lead to disengagement of individual components and between the core and interfacing structures and to a loss of control of the core geometry, graphite growth could lead to the take-up of design clearances and large forces between structures, and differential shrinkage/growth in a component could lead to high stresses and failure by cracking.

b) Oxidation

The graphite oxidation work involves two principal tasks relevant to safety analysis and licensing of HTRs for normal operation.

- The improvement of experimental database for advanced graphite oxidation models.
- Experimental investigation of innovative C-based materials for application on HTRs.

Graphite burning under severe air ingress accidents, is assessed by computer models, based (up to now) on isothermally measured kinetic equations which consider in-pore diffusion and chemical reaction mechanisms. The data requirement for such models are burn off dependent chemical (regime 1) reactivities and in-pore diffusion coefficients. The experimental work to be undertaken deals with the case of severe air ingress, with the diffusion influences of the oxygenation process determined using the thermo-gravimetric facility THERA housed at FZJ. Measurements are made for the German A3 fuel matrix graphite (type A3-27) and for a typical structural graphite (V483T fine grain graphite) in oxygen at temperatures between 823 and 1023 K and oxygen partial pressures between 2 and 20 kPa.

Although material development for HTRs almost ceased in Germany in 1985 development of C-based materials for Fusion Reactors continued. These included innovative concepts such as CFCs, and mixed materials (Si, Ti content) which show a high heat conduction and high strength. A second series of experiments are therefore planned to look at the performance of such materials under accident typical temperatures (approximately 1273 K) in steam and in air. The experimental work involves the use of induction heated tube shaped samples in an induction furnace facility. Oxidised gas flows 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 are measured by mass spectrometric analysis of the product gases and by weight loss of the sample. Experiments are also planned for measurements of CFC materials irradiated and non-irradiated (up to 1dpa) that will look at strength, dimensional change, thermal diffusivity and heat capacity.

HTR Materials Database

The need for reliable material data and properties is a key issue in the development of any innovative reactor technology and is especially important for the understanding of structural integrity issues and material behaviour. It is particularly important in those areas where safety considerations are uppermost. For the HTR key component materials a database of properties is required that covers information on manufacturing provisions, products & parts, test data information and design properties. Reliable design property information is one of the main outputs and needs. Work on developing a database is underway with considerations being given to the development of a web-based facility. Design and technological issues plus omissions and shortage of test data will be the basis on which the database will be built up and expanded and on which future R & D needs will be assessed.

Summary and Conclusions

The need for reliable material data and properties is a key issue in the development of HTR technology and is especially important for those areas where safety considerations are uppermost. The HTR-M & M1 projects aim to provide a firm materials platform on which to develop the future HTR technological basis within Europe and provide a first step towards building an understanding of the behaviour and manufacturing requirements for some of the new materials that will be needed for such developments. The objective is to improve knowledge of materials for future HTRs for the reactor pressure vessel, the control rod and turbine (blades & discs) and the reactor graphite. The projects extend over a period of four years and are being performed with the support of the European Union Fifth Framework Programme. The project is also being co-ordinated through the direction of a European HTR Technology Network (HTR-TN). This paper outlines the Work Programmes and reviews important issues concerning material needs for feasibility and assurance of structural integrity.

Acknowledgements

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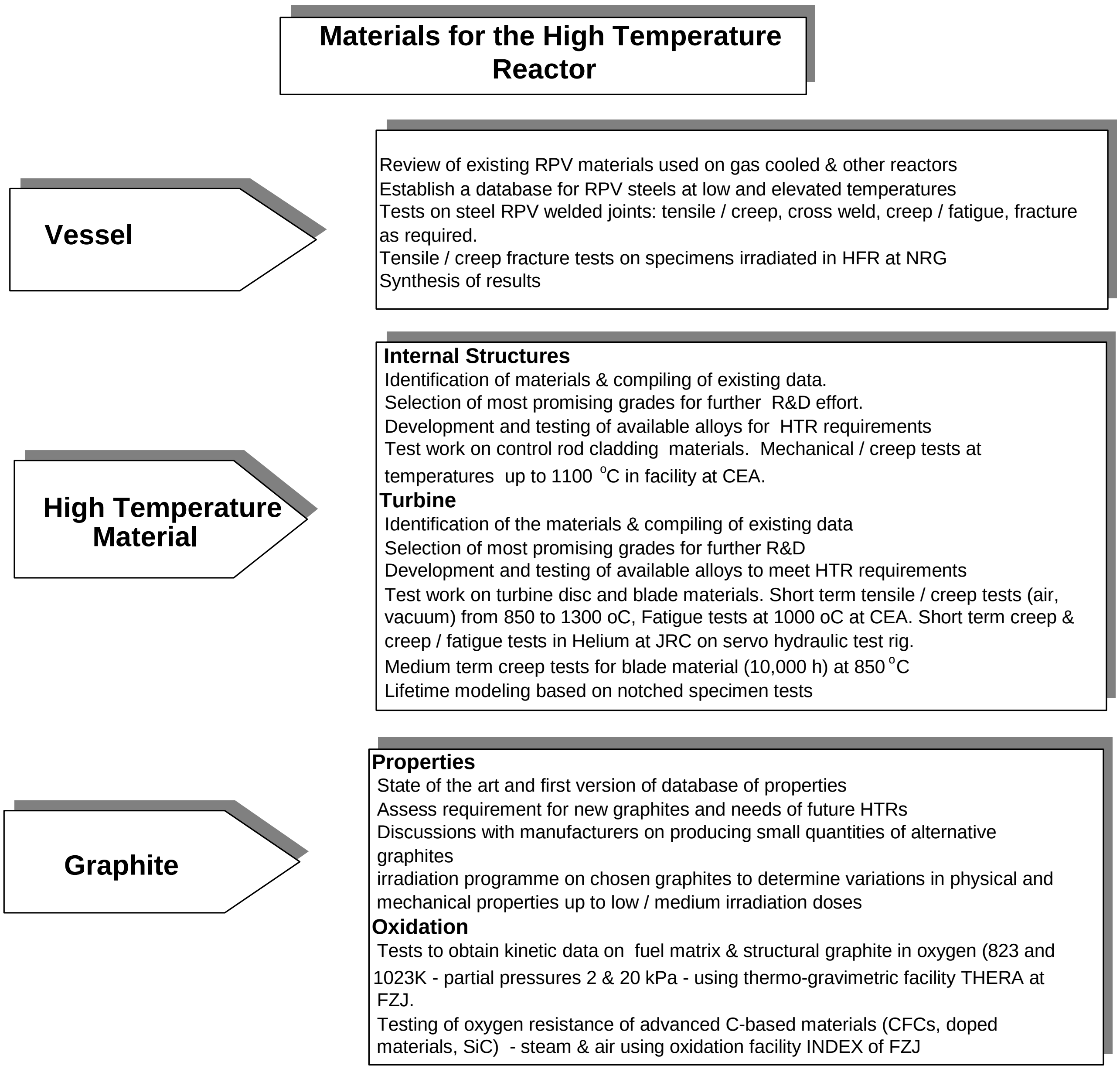

Figure 1 Summary of HTR-M & HTR-M1 Work Programs

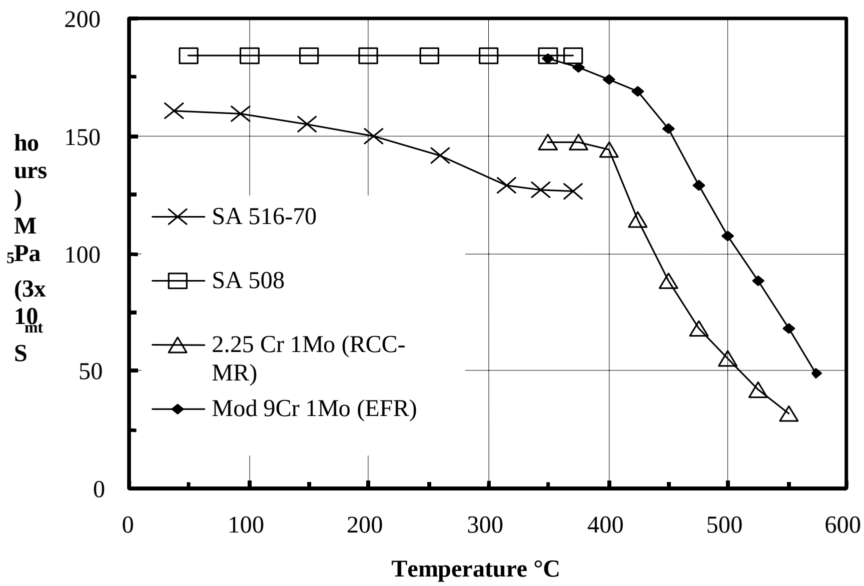

Figure 2 Allowable Stresses For RPV Materials

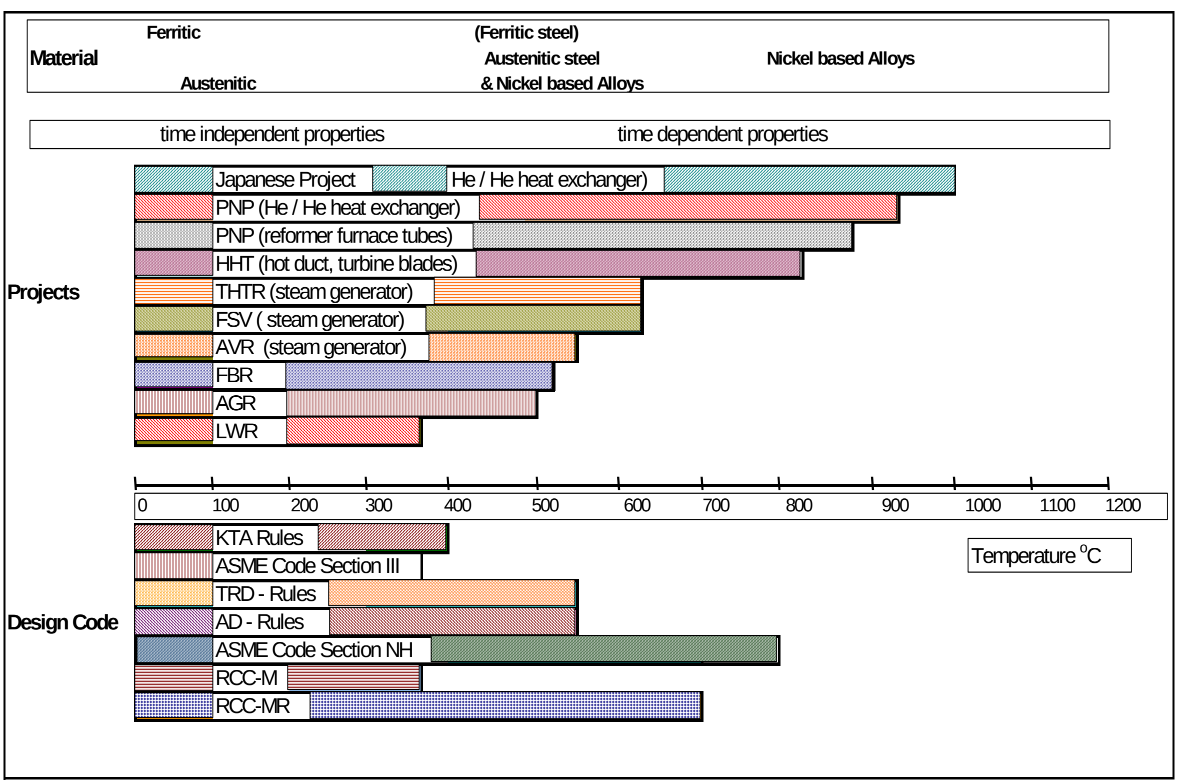

Figure 3 Materials Temperature versus Design Code Temperatures