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State of the art review of HTR vessel materials.

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UK

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Table of Contents

List of Tables

List of Figures

Summary

The state of the art with respect to materials for HTR vessels is outlined based on HTR-M studies covering materials selection, design issues, design codes, materials data, manufacturing, irradiation effects and thermal ageing effects.

The ‘cold’ vessel design concept adopted for the PBMR reactor pressure vessel leads to the choice of Mn-Ni-Mo or C-Mn steel with grades similar to SA 508 Class 2 as prime candidates. Advantage can be taken of PWR vessel technology and design rules.

The ‘warm’ vessel concept adopted for the GT-MHR reactor pressure vessel leads to the choice of Cr-Mo steels with modified 9Cr-1Mo steel grades similar to ASME Grade 91 as prime candidates.

It is concluded that the HTR-M project should support both concepts by a combination of experimental work to supply data on the irradiation and thermal ageing resistance of modified 9Cr-1Mo steel and the compilation of existing data covering all candidate materials.

Future actions covering the provision of design materials data and manufacturing technology are identified. The HTR-M project can cover most of the required actions but work in the areas of manufacturing development and long-term creep rupture testing is beyond the resources of current projects within the HTR cluster.

1 Introduction

The HTR-M project has as one of its main objectives the improvement of reactor pressure vessel and other pressure vessels that contain the components of a direct cycle HTR. It is anticipated that improvements will be found on the basis of progress in the related areas of design analysis, structural integrity analysis and the provision of materials data covering mechanical properties, irradiation effects and compatibility with the coolant.

2 The HTR-M Project

The objective of the HTR-M project is to provide materials data for key components of the development of High Temperature Reactor technology in Europe. The need for reliable materials data and properties is a major issue in the development of any innovative reactor technology and is especially important for those areas where safety considerations are uppermost. The aims of this project are:

- to provide a firm materials platform on which to develop the future HTR technological basis within Europe and;
- to 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 work programme is split into three Work-Packages (WP 1-3) according to material type and the conditions under which they operate.

WP1 covers this review of vessel materials, the setting up of a materials property database and an R&D programme covering the manufacture of a welded joint in a selected material for tensile, creep and fracture toughness testing, including the effects of irradiation.

In WP2 consideration is given to the high temperature regions of the reactor covering the (WP2.1) internal structures (i.e. control rod) and (WP2.2) the turbine (discs and blades).

WP3 involves setting up the database of properties on graphite. The aim is to retain existing experience from gas cooled and high temperature reactors.

3 Scope of the review

This review covers pressure vessel materials for modern HTR concepts. These involve systems having inherently safe characteristics with potentially significant advantages with respect to simplicity and public acceptance. A fundamental part of these systems is a Reactor Pressure Vessel (RPV) made from steel and able to transmit stored and decay heat to its surroundings at a sufficient rate to maintain acceptable transient temperatures within the vessel (including peak core temperatures) under the most adverse circumstances.

Modular power generation systems with gas turbines are economically attractive due to the consequent elimination of complex safety systems. The modular systems involve another large pressure vessel containing the power conversion system (PCSV). A ‘cross vessel’ (XV) of smaller diameter connects the RPV to the PCSV.

Other potential applications of HTRs retain RPV made from steel but may need other vessels with different characteristics to meet requirements such as:

- combined electricity generation and process heat for desalination plants;
- high temperature process heat for hydrogen and synthetic fuel production.

This review concentrates on candidate materials for the RPV. Requirements for the PCSV and XV are also considered where information is available and there are potential implications for either the fabrication or service conditions of the RPV material.

The review relies heavily on work done within the HTR-M project (Refs. 1-9) by experts in the fields of vessel design, structural analysis, materials science and manufacturing technology. In order to provide a concise account much of the detail in Refs. 1-9 has been omitted. The full documents are available on the SINTER website but summaries of their scope and conclusions are included in this review.

In subsequent sections the state of the art with respect to materials and design aspects of the RPV are reviewed as follows.

1. A short list of candidate materials is presented and some basic design considerations are identified on the basis of a survey of existing and proposed HTRs.

2. The RPV design background to the HTR-M project is considered in terms of the anticipated vessel geometry and loading conditions. The use of existing design codes is outlined along with damage mechanisms that potentially arise over the lifetimes of HTR pressure vessels.

3. Candidate material grades in design codes are then listed and compared in terms of their design code status, their design materials properties as well as the availability of manufacturing technology and experience.

4. Environmental considerations including the effects of irradiation, thermal ageing and gas-metal reactions are discussed.

It is shown how these considerations led to the choice of modified 9Cr-1Mo steel for the vessel manufacturing and materials property R&D programme of the HTR-M project. The need for work on other candidate materials is also discussed with emphasis on building up the HTR-M database on vessel materials.

4 Pressure vessels of existing and proposed HTRs

Data on test reactors and various designs for future HTRs were reviewed in Ref. 1. A list of relevant reactors and design data was compiled based on data collected in an IAEA document (Ref. 2) and additional information provided by JAERI (Ref. 3). Key data are given in Table 1 including the design pressure and vessel diameter, which dominate the choice of material and the vessel thickness.

The test reactors, HTTR and HTR 10, have relatively low vessel diameters and internal pressures compared with the commercial designs. Pressure vessels of lower thickness and materials with lower strength are therefore acceptable. In the commercial designs vessel diameters and internal pressures are somewhat greater but fall within relatively narrow ranges. This is the result of constraints imposed by a combination of passive cooling requirements and the design strength of candidate materials.

Four types of steel are identified in Table 1, C-Mn steel is specified for HTR-10 while a comparatively low strength grade of 2¼Cr-1Mo steel is specified for HTTR. The commercial reactors fall into two groups depending on the RPV operating temperature. A particular material type is associated with each vessel group.

- Mn-Ni-Mo steel (ASME SA508 or equivalent) is specified for 'cold' vessels operating at less than 350°C. In this group HTR-M participants are most interested in the South African PBMR system.
- Modified 9Cr-1Mo steel (ASME Grade 91 or equivalent) is specified for 'warm' vessels operating at up to 490°C. In this category HTR-M participants are most interested in the GT-MHR system designed in Russia with support from the USA, Europe and Japan.

The existence of 'cold' and 'warm' vessel options is of great significance to the work of HTR-M because resource limitations dictate that a single material has to be chosen for the R&D programme including the planned irradiation experiment. This point will be considered later in the light of the design and materials studies discussed below.

5 Design background and design codes

5.1 Classical Design methods and codes.

HTR-M studies in the areas of vessel design and design codes (Refs. 2-6) have attempted to determine the significance for HTR RPVs of the five classical 'damage mechanisms' (DMs):

DM 1. Instantaneous excessive deformation.
DM 2. Delayed excessive deformation.
DM 3. Ratchetting.
DM 4. Fatigue
DM 5. Creep-fatigue.

DMs 1 and 2 are of major significance to the design of pressure vessels because they lead directly to the primary stress limits S_m (instantaneous limit based on tensile properties) and S_t (delayed limit based on creep properties). It will be seen later that the crossover temperature θ_{co} above which S_t applies is an important parameter for RPV materials selection.

Ref. 5 identifies sensitive zones of the vessel where the other damage mechanisms (DMs 3-5) may have the most effect. Although Refs. 2 and 4 give some indications of temperatures and pressures during normal operation and transients it has not been possible with the state of knowledge available currently within HTR-M to make detailed analyses. Ref.5 lists sites of potential damage but gives no indications of the temperatures and stresses involved. Items relevant to DMs 3-5 are identified below.

- Parts of the vessel where wall temperatures are highest may be subject to creep damage.
- Nozzles and openings including the cross-vessel connection and penetrations in the hemispherical bottom and lid may be subject to fatigue.
- The internal support attachments may be subject to fatigue and ratchetting.
- The RPV and PCU support to foundation may be subject to fatigue with mean stress effects. The designs of the two supports need to be compatible.
- Welds.

In Ref. 6 the application of the RCC-MR and ASME Codes to the design of HTR RPVs is described in detail. The need for improvements and verifications of the codes are identified as well as missing materials data.

Section III, Division 1, Subsection NB of the ASME code is fully applicable to the design of ‘cold’ HTR pressure vessels operating at up to 371°C. Although not considered in Ref. 6 the RCC-M Code provides a European set of rules for ‘cold’ vessels. Both ASME III NB and RCC-M incorporate extensive LWR experience and are easily available. They can be regarded as having equivalent status from the standpoint of a cold HTR vessel.

The ASME III Division 1, Subsection NH and the RCC-MR rules (including changes/additions to the next issue) provide limited coverage for ‘warm’ HTR vessels.

ASME III Division 1, Subsection NH has not been updated for many years and currently covers only 2 ¼ Cr-1Mo steel.

The RCC-MR rules were written for liquid metal cooled fast reactors. Their application to HTR vessels requires that the following issues be addressed.

- It will be necessary to check the applicability of existing ratchetting rules to a cyclic softening material such as modified 9Cr-1Mo steel and to provide relevant data.

- The effect of high levels of mean stress in fatigue under HTR conditions needs to be incorporated.
- It will be necessary to obtain data for creep-fatigue assessment (mainly from other codes and documents) to supplement RCC-MR data mainly for 2 ¼ Cr steel.
- A negligible creep curve for mod 9Cr steel may be required.
- Weld joint coefficients for relevant materials and conditions are not available in RCC-MR.

Ref. 6 notes that modifications have been developed to classical design rules in order to allow for the effects of irradiation on some components. Irradiation conditions at an HTR vessel wall are broadly similar to those in PWR vessels, i.e. neutron energies and fluences are such that damage levels are well below 1dpa. At these levels irradiation can have a significant effect on the fracture toughness of pressure vessel steels (see 6 and 7 below) but a common position exists in LWR and LMFBR classical design rules.

- Irradiation induced effects (swelling, irradiation induced creep) have no significant effect on stress levels or stress distributions.
- Ductility and strain hardening characteristics will not be affected significantly.
- The minimum yield and ultimate tensile strength of the as fabricated base material and weld metal provides a lower bound to the values after irradiation. No credit is taken for any increases in yield strength, UTS or S_m due to irradiation.

The only change to the classical design rules for irradiation to less than 1 dpa concerns austenitic stainless steels when creep is significant. Irradiation may produce sufficient He in these materials to reduce creep strength, (S_r and S_t) and increase the creep damage component in creep-fatigue. This effect is not significant in the ferritic steels under consideration for HTR pressure vessels.

5.2 Flaw tolerance

Flaw tolerance analyses are necessary in addition to classical design rules to assess pressure vessel integrity at both the design stage and during its service life. Both the RCC-M and ASME design codes provide bases for various types of analyses including:

- verification of resistance to fast rupture from reference defects, realistic defects in relation to fabrication/inspection methods and actual defects detected during in-service inspection;
- demonstration of leak before break capability.

The following materials data are required for base materials and welds in addition to the tensile properties used in classical design rules.

- Fracture toughness vs. temperature.
- Fatigue crack growth data with allowance for creep and environmental interactions.

It is also necessary to establish the effects of irradiation and thermal ageing on the above properties.

The availability of relevant data is considered in more detail in Sections 6 and 7 below.

6 Candidate Material Grades

6.1 Available Grades

A survey of the steel grades corresponding to the four types of material identified in Ref. 1 above is reported in Ref. 7. Table 2 identifies the grades in the ASME code (based on ASTM standards), in the RCC-MR code and/or AFNOR standards as well as the Russian code (Ref. 12) or GT-MHR design documents (Refs. 13 and 14).

As noted above the codes provide properties for many materials only up to temperatures of 370-375°C. Ref. 7 therefore identifies two additional sources of data:

- code cases that contain additional design material data;
- non-code documents containing compilations of data that can be used to derive design materials data.

Suitable documents and the corresponding temperature limits are listed in Table 3.

Definitive transient temperatures for emergency/accident conditions are not available to HTR-M participants at present but Table 3 shows that tensile and creep data are available for temperatures up to 200°C above the limits for normal operation. A basis therefore exists for determining primary stress limits under accident and emergency conditions.

6.2 Materials properties for classical design

Comparisons between material types and between grades of the same material type are given in Ref. 8 for the following cases.

- ASME Grade 91 and RCC-MR mod 9Cr-1MoVNb.
- Various grades of 2 ¼ Cr-1Mo steel (ASME Grade 22 Class 1, the normalised or quenched and tempered grades of RCC-MR.
- Mn-Ni-Mo steel (PWR grades).

Comparisons of tensile and creep data for matching grades indicate that there are differences in the minimum stress to rupture S_r and the primary stress intensity S_t, particularly for modified 9Cr1Mo steel. There is a need for long-term creep data at moderate creep temperatures (425-500°C) to confirm long term values of S_r and S_t for both the 2 ¼ Cr-1Mo and modified 9Cr-1Mo types of steel.

In order to minimise the impact of the uncertainties associated with S_r and S_t, Ref. 8 recommended that each grade should have a normal service temperature lower than θ_{CO} so that the time independent primary stress limit S_m is less than S_t. A similar recommendation was made in Ref. 3 on the grounds that allowable stresses fall rapidly with increasing temperature above θ_{CO}. Values of the crossover temperatures for a service life of 60 years according to Ref. 8 are:

- 494°C for ASME Grade 91;
- 435°C for the RCC-MR mod 9Cr-1MoVNb grade;
- 420°C for 2 ¼ Cr-1Mo to ASME Grade 22 Class 1;
- 410°C for the normalised or quenched and tempered grades of RCC-MR;
- 371°C for the Mn-Mo-Ni grades.

Compared with modified 9Cr-1Mo the minimum vessel thickness for a given diameter and internal pressure has to be increased by the following factors if different materials are used.

- 1.25 for the normalised or quenched and tempered grades of RCC-MR.
- 1.5 for 2 ¼ Cr-1Mo to ASME Grade 22 Class 1.

No increase is necessary for the Mn-Mo-Ni grades but normal service and transient temperatures are limited to 371°C.

The differences in the creep strength of modified 9Cr-1Mo steel according to ASME and RCC-MR have a significant effect on θ_{CO}. The GT-MHR reactor pressure vessel design temperature of 440°C is above θ_{CO} according to RCC-MR but 50°C below θ_{CO} according ASME.

It should be noted that when modified 9Cr-1Mo was introduced into the RCC-MR rules a change was made to one of four the criteria used for ferritic steels to obtain S_m from the tensile properties. The criterion based on the minimum tensile strength at elevated temperature is R_m (θ°C)/F. The value of F for modified 9Cr-1Mo steel was changed from 3 to 2.7 to ensure that S_m values matched those proposed for use in the ASME Code. This 10% increase in S_m reduces θ_{CO} for a 60 year life from 555° to 535°C.

In Ref. 8 a comparison was made of the design fatigue curves and creep-fatigue interaction diagrams given in ASME and RCC-MR for 2 ¼ Cr-1Mo and modified 9Cr-1Mo steel. It was concluded that fatigue and creep-fatigue properties were not important criteria for pressure vessel materials selection. A definitive conclusion on this question would need to take into account representative cycles for one or more sensitive zones of an HTR vessel. The required sets of local stresses, strains and temperatures based on the rules in appropriate design codes are not available to HTR-M participants at present.

6.3 Materials properties for flaw tolerance assessments

Fracture toughness and fatigue crack growth data excluding the effects of irradiation and thermal ageing are provided in LWR design codes such as ASME and RCC-M for Mn-Ni-Mo steel plate, forgings and welds. In the case of fracture toughness the codes provides two sets of reference data.

- Toughness in the transition regime as a function of temperature related to a reference temperature (RT_{NDT}) that has to determined from drop weight and Charpy impact tests on the base material and/or weld metal.
- Upper shelf toughness as a function of temperature.

These relationships will be fully applicable to a ‘cold’ HTR vessel at the start of life provided that the following conditions are met.

- Mn-Ni-Mo steel is specified.
- The full requirements of the LWR codes are followed during material procurement, manufacture and testing, including full provision of data to allow determination of RT_{NDT}.

In principle the same approach can be adopted for warm vessels manufactured from 2¼Cr-1Mo or modified 9Cr-1Mo steel. The current situation with respect to data provision is set out in Ref. 8.

In the case of 2¼Cr-1Mo steel detailed toughness characterisations have been performed for a Japanese grade similar to ASME SA 387 Class 2 (Ref. 25) and a French grade similar to ASME SA 541 Grade 22 Class 3 (Refs. 26 and 27). Provided that low impurity levels (S and P) are maintained both grades have low transition temperatures and high upper shelf toughness. The reference toughness curve of ASME III Appendix G was considered appropriate for the French grade but a modified definition of the reference temperature was proposed in Refs. 26 and 27.

During development of the modified 9Cr-1Mo steel for LMFBR steam generator tube plates it was shown that the forged and heat-treated material had high toughness throughout the thickness of the plate. The resulting RCC-MR procurement specifications call for higher impact toughness values compared with 2¼Cr-1Mo steel forgings. The material was not fully characterised by impact, drop weight and fracture toughness testing. It is not possible at this stage propose a procedure for reference temperature determination or to validate the use of the ASME III Appendix G reference transition curve.

6.4 Manufacturing considerations

6.4.1 Vessel manufacturing route

The technology developed for building PWR vessels from heavy section ring forgings provides a sound basis for ‘cold’ vessel manufacture covering:

- procurement specifications for forgings;
- welding procedures and filler metal selection;
- manufacturing sequences including weld examinations and post weld heat treatment.

HTR vessel wall thicknesses are within the PWR range but the diameters of HTR vessels (6-8.5 m) are much greater than for PWR (up to 4.5m). There is therefore a need to establish a capability to produce forged parts of the required dimensions having the required mechanical properties. If this cannot be done it will be necessary to manufacture the vessel from formed plates. In this case there are implications for both design and manufacturing. The most important design consideration is the introduction of axial welds with higher cross-weld stresses. Manufacturing requirements include provision of procurement specifications, forming facilities and forming procedures for heavy plates.

On-site welding and post weld heat treatment (PWHT) of the Reactor Vessel-Cross Vessel- PCS Vessel assembly introduces a requirement that does not arise in PWR manufacture. It will be necessary to develop one of the following:

- a validated local heat treatment procedures for Mn-Ni-Mo steel;
- a site heat treatment facility that can accommodate the whole Reactor Vessel-Cross Vessel- PCS Vessel assembly.

The above considerations also apply to ‘warm’ vessels. In the case of 2¼ Cr-1Mo steel, Ref. 8 reports that there is manufacturing experience in France for one grade and it can be extended to other grades of 2¼Cr-1Mo steel. Appropriate filler metals exist but the properties of welded joints require evaluation. Russian experience of WWER-1000 vessel manufacture in 2¼Cr-Ni-Mo-V steel is also relevant. The PWHT temperature for 2¼Cr-1Mo steel is 630-660°C compared with 620°C for Mn-Ni-Mo steel. This difference may have a significant effect on cost particularly in the case of a site PWHT facility.

In the case of modified 9Cr-1Mo steel the above noted manufacturing problems introduce greater difficulties and costs associated with:

- a lower limit on the mass of single cast or ingot for high Cr steel compared with 2¼Cr-1Mo steel;
- difficulties in selecting filler metals to achieve the required levels of resistance to hot cracking, high toughness at 0°C and creep strength;
- a much higher PWHT temperature (>750°C) compared with 2¼Cr-1Mo steel (630-660°C). Local heat treatment at >750°C of welds in the RPV/cross vessel assembly would be particularly difficult to validate;
- the design temperature of the PCS vessel for GT-MHR is 150°C compared with 440°C for the RPV and cross vessel. It is possible therefore to specify C-Mn, Mn-Ni-Mo or 2¼Cr-1Mo steel for the PCS vessel. A transition joint between modified 9Cr-1Mo steel and any one of the other candidate material types would involve a mismatch of 100-150°C in their PWHT temperatures. It might be necessary in this case to validate the use of a filler metal such as Alloy 82 and a ‘buttering’ technique

to permit separate PWHT at temperatures appropriate to each base material prior to completion of the closure weld.

6.4.2 Properties of welded joints

For the Mn-Ni-Mo steel welds in ‘cold’ vessels PWR experience provides filler metals and welding procedures giving toughness properties well matched to the base material. Because some transients may involve temperatures exceeding θ_{co}, cross weld creep tests are required to determine whether reduction factors need to be applied to the relatively short time-high temperature design data appropriate to the assessment of transients.

For ‘warm’ vessels, Ref. 8 indicates that the start of life toughness of the 2¼Cr-1Mo steel HAZ and weld metal match that of the base material. The same is true for the HAZ of modified 9Cr-1Mo steel but adequate weld metal toughness depends on the solution of the filler metal selection problem mentioned in 6.4.1.

Ref. 8 quotes values of the creep strength reduction factor J_r from ASME III NH for 2¼Cr-1Mo steel to ASME Grade 22 Class 1 used with selected filler metals from American Welding Society (AWS) standards. There is no reduction for temperatures up to 454°C and a very small reduction at 482°C (J_r = 0.99 for 3x10^5 h).

Values of J_r for modified 9Cr-1Mo steel with matching filler metal are controlled by the strength of the HAZ. In tensile and short term cross weld creep tests at lower temperatures the coarse grained HAZ controls failure. In long term tests at higher temperatures cross weld strength is limited by higher creep rates in the fine grained ‘intercritically annealed’ HAZ leading to cavitation and ‘Type IV’ cracking. Type IV cracking has been observed at 600°C and 650°C but not in tests at 550°C extending to 20000 h. These trends are taken into account in the recommended values of Ref. 21 shown in Table 4. These values are preliminary lower bound estimates based on limited data.

A large programme of short and long term cross weld tests would be required to provide full coverage in design codes at temperatures of 400-550°C for both 2¼Cr-1Mo and modified 9Cr-1Mo steel. The following more restricted requirements for ‘warm’ HTR vessels can be proposed.

- Cross weld creep tests at 400-450°C on 2¼Cr-1Mo steel to specifications other than ASME Grade 22 Class 1to justify the use of J_r = 1 for long term service temperatures of 410-420°C.
- Long-term creep tests at 400-500°C on modified 9Cr-1Mo steel to establish time dependent J_r values at temperatures below θ_{co} (435/ 495°C according to RCC-MR/ASME).
- Assessment of collected data to establish short-term J_r values, at temperatures up to 550°C, for the assessment of possible transients in ‘warm’ vessels.

7 Environmental Effects

Potential environmental effects identified in Refs. 1 - 9 include the effects of irradiation and thermal ageing in addition to corrosion effects arising from interactions between the pressure vessel steel and impurities in the He coolant.

7.1 Irradiation embrittlement

7.1.1 Cold vessel materials

The effects of irradiation on the toughness of LWR steels have been studied extensively and reviews have been undertaken as part of the work programme of the CEC supported AMES network (**A**geing, **M**aterials **E**valuation and **S**tudies). Reports from this group (Refs. 29 and 30) as well as publications in the open literature (Ref. 30) are highly relevant to the materials and operating conditions of 'cold' HTR vessels given that LWR vessels operate at 250-300°C with similar end of life fluences to HTR vessels.

The major effect in these materials is an increase in the DBTT (ΔDBTT) due to the combined effects of matrix hardening and grain boundary weakening associated mainly with the residual elements P and Cu.

Various empirical relationships between composition, fluence and ΔDBTT have been proposed on the basis of results from test reactors and LWR surveillance programmes (Western PWRs and Russian LWRs. These were reviewed in Refs. 29-31. Common features of the formulae are the importance of the Cu and P contents as well as synergistic effects of the Ni content. The range of behaviour predicted for Mn-Ni-Mo, 2¾Cr-Mo-V and 2¼Cr-Ni-Mo-V pressure vessel steels is shown in Figure 1. The upper bounds correspond to steels used in the 1960's and 1970's which had relatively high levels of either Cu (PWR steels) or P (WWER steels). Specifications for modern pressure vessel steels and weld metal contain special limits on Cu and P for material to be used in the most highly irradiated (beltline) region of LWRs as follows:

- 0.008%P and 0.08%Cu in RCC-M;
- 0.012%P and 0.12% Cu in the ASME Code;
- 0.010% P and 0.08% Cu in WWER rules.

The above limits can be met comfortably using modern steelmaking techniques. They ensure that the predicted end of life ΔDBTT for a LWR vessel operating at 250-300°C is small in relation to the data scatter.

These findings and the resulting improvements to pressure vessel steel specifications can be applied directly to 'cold' HTR vessels because steel grades, vessel dimensions, operating temperatures, end of life fluences and neutron spectra match those in LWR vessels.

7.1.2 Warm vessel materials

Ref. 9 describes a review of published data covering the effects of irradiation and thermal ageing on tensile properties and toughness. The review was intended to support the planning of the HTR-M irradiation experiment on a thick section modified 9Cr-1Mo welded joint. Because data on modified 9Cr-1Mo steel were relatively sparse, steels containing 2¼ -12% Cr and irradiation temperatures of 250-500°C were considered. The effect of irradiation level on ΔDBTT is shown Figure 1.

Inspection of Figure 1 shows that no data were available for modified 9Cr-1Mo steel irradiated under the conditions proposed for the HTR-M test programme in HFR (8 mdpa, 400-460°C). All of the data for modified 9Cr-1Mo steel were obtained at much higher doses (≥0.8 dpa) though low dose data were available for other steels. Ref. 9 concludes that:

- there is a strong effect of irradiation temperature in the range 250-450°C;
- at 250-300°C irradiation effects in modified 2¼ -12% Cr steels will be similar to those in LWR steels with low levels of P and Cu;
- at GT-MHR operating temperatures (400-440°C) irradiation effects on ΔDBTT will be very small, possibly negligible;
- irradiation at 350°C would probably give a measurable effect;
- limited data on other high Cr steels indicate that the effects of irradiation on weld metal and HAZ material are similar to those on parent metal.

In Ref. 8 it is argued that irradiation effects in Cr steels are expected to be lower than in Mn-Ni-Mo steel due to the reduced Ni content of 0.2-0.3% in the Cr steels. Data for 2¼Cr-1Mo steel to SA 541 Grade 22 Class 3 (presumably with low Cu and P contents) indicate that ΔDBTT was less than 20°C and the upper shelf Charpy energy was high (>250J) after irradiation to 3.5 x 10^{23} n/m^2. Ref. 8 also mentions severe reductions in upper shelf energy in modified 9Cr-1Mo steel irradiated to 30 dpa. Similar effects are included in Figure 2 but the dose is very high when compared with end of life fluences for PWR and HTR vessels and the most severe effects occur at low temperatures.

7.2 Thermal ageing embrittlement

7.2.1 Background

Thermal ageing embrittlement (TAE), sometimes called temper embrittlement (TE), may involve hardening due to precipitation of carbides and intermetallic compounds along with grain boundary segregation of trace elements (P, Sn, As, Sb). The term temper embrittlement came into use because slow cooling after tempering was sufficient to embrittle steels with relatively high levels of the trace elements. More severe effects could be induced by step cooling from the tempering temperature followed by holding at approximately 450°C and or slow (furnace) cooling to room temperature.

TE is not usually a problem in modern steels with very low levels of P and the other trace elements. TAE can however occur after long-term service at temperatures in the range 350-550°C. The severity of the effect depends on a number of factors including:

- the prior austenite grain size of the material;
- the concentration and distribution of the trace elements P, Sn, As and Sb;
- the cooling cycle after tempering;
- time at temperature(s) within the critical range;
- the property considered.

The base materials and weld metals of both cold and warm HTR vessel steels normally have very fine grain sizes. The most likely embrittlement location is therefore the coarse grained heat affected zone. This is a narrow region adjacent to the fusion boundary where temperatures of 1100-1300°C are reached during welding.

Cold vessels operate at temperatures below the embrittlement range so TAE can occur only during transients involving exposure to higher temperatures for tens or hundreds of hours. Warm vessels operate in the TAE temperature range throughout their lifetime and the possible effect of TAE of on vessel integrity is of greater concern. The two cases are considered separately below.

7.2.2 Cold vessels

LWR studies (Refs. 32 and 33) have concentrated on pressure vessel materials heat treated to simulate coarse grained heat affected zones and aged at 400-490°C. This is consistent with the view expressed in Refs. 8 and 28 that long-term thermal ageing of base material and weld metal under normal operating conditions does not produce significant changes in tensile properties or toughness.

Work on laboratory produced heats of steel with typical RPV compositions (Ref. 32) showed that severe embrittlement could be induced in simulated worst case weld HAZ microstructures having very coarse prior austenite grain sizes (ASTM 0-1). These were produced by oil quenching from 1200°C followed by post weld heat treatment at 615°C for 24 hours with oil quenching to maximise the potential for subsequent ageing embrittlement. In the ‘fully aged’ condition (2000 h at 450°C), ΔDBTTs of 118-209°C were found. The cast having a ΔDBTT of 118-209°C after ageing showed a shift of only 69°C after irradiation to 12.5mdpa at 288°C. The P and Cu levels in this cast (0.017% and 0.16% respectively) were deemed ‘high’ in Ref. 32 but they are relatively low by historical PWR and/or WWER standards.

In subsequent studies (Ref. 33), various heat treatment and ageing cycles were applied to five commercially produced heats of RPV steel having P and Cu levels similar to the laboratory materials of Ref. 32.

The heat treatment of Ref. 32 produced prior austenite grain sizes of ASTM 0-00. In the ‘fully aged’ condition of 2000 h at 450°C DBTT shifts of 35-145°C were obtained i.e. significantly lower than in the Ref. 24. An ageing treatment of 168h at 490°C, corresponding to the maximum irradiation damage annealing cycle, produced a similar range of DBTT shifts (34-132 °C).

More precise simulations of the welding thermal cycle were applied to the commercial steels in a ‘Gleeble’ fast heating and cooling facility. Grain sizes of ASTM 4-5 were produced and ΔDBTT values for fully aged material ranged between 56-133°C.

All of the above results were obtained under conditions of prior heat treatment that favoured thermal ageing embrittlement (coarse grain sizes, oil quenching after tempering and long term ageing at 450°C. It is likely that in real welds HAZ toughness will be less severely affected. The data do however serve to illustrate the need to take precautions in the preparation of procurement specifications and during manufacture covering:

- control of concentrations of critical elements;
- avoidance of very coarse HAZ grain sizes;
- control of cooling rates after tempering

7.2.3 Warm vessels

Refs. 8 and 9 consider the effects of thermal ageing on base material. Figure 2 shows the effects on the DBTT and Upper Shelf Energy (USE) of 9%Cr steels as presented in Ref. 9. The data are sparse and the time-temperature parameter in Figure 4 is based on tempering rather than ageing behaviour. Despite these restrictions it can be concluded as in Ref. 9 that the effect of thermal ageing over the lifetime of the HTR vessel (400,000 hours at 400-460°C) on the DBTT and USE of wrought modified 9Cr steel would be small. However no data were available for the effect of thermal ageing on modified 9Cr welds. In view of the observations of significant thermal ageing embrittlement in the coarse-grained HAZs of LWR steels it was recommended that the effect of ageing on modified 9Cr welds be investigated.

In Ref. 8 it was noted that TE was an old problem in 2¼Cr-1Mo steel but the shift in DBTT would be low at HTR operating temperatures and low P+Sn concentrations. It was noted that the effect of thermal ageing on USE required further evaluation, particularly for weld metal.

7.3 Gas-Metal Reactions

Design codes usually instruct the designer to take into consideration the effects of corrosion without specifying in detail how this should be done.

In practice two measures are usually adopted.

- An allowance for metal losses over its lifetime is added to the minimum design thickness of a pressure vessel.
- Fatigue and creep-fatigue damage calculations made with design data include allowances for data scatter, ‘mild’ (oxidising) environments and the characteristics of typical pressure vessel surfaces. Additional margins are necessary only where the environment has different and/or more severe effects on fatigue and creep e.g. by carburising exposed surfaces or by producing intergranular oxidation to a significant depth.

In Ref. 1 it was noted that HTR gas chemistry is dominated by the impurities H_2, H_2O, CO, CO_2. Three factors control the He impurity levels:

- the hydrogen and oxygen ingress rates;
- the core temperature and gas residence time, the latter is usually considered to be sufficient for equilibrium to be achieved at 850 - 1000°C;
- the capacity of the gas clean-up plant.

Thermodynamic assessments of five selected HTR atmospheres in terms of the carbon activity and oxygen potential (notional partial pressure of oxygen) showed that:

- all five atmospheres have carbon activities greater than unity at 300 and 350°C i.e. they will tend to deposit carbon;
- all five atmospheres are either neutral or carburising with respect to Fe/ Fe_3C., carburisation of C-Mn steel or SA 508 type material is therefore the main risk for cold wall vessels;
- in order for breakaway oxidation (as in CO_2 based AGR atmospheres) would arise in He only if the water content was increased by approximately two orders of magnitude, such conditions are unlikely to arise in a direct cycle system and would anyway be unacceptable in the long term for other reasons such as graphite oxidation;
- all five atmospheres were either neutral with respect to the equilibria between Fe, Fe_3C and Fe_3O_4 or well within the field where Fe is the stable phase, the main risk to a ferritic or martensitic steel was therefore decarburisation, particularly at 540°C;
- chromium and its carbides will be oxidised in all five gas compositions, when the gas composition is also neutral or stable with respect to Fe there is a risk of internal oxidation of Cr and also more reactive elements such as V and Nb modified 9Cr-1Mo steel.

It therefore appears that HTR metal losses will be 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. Given a beltline wall thickness in the range 140-200 mm metal loss will only become significant if unstable processes such as breakaway corrosion or metal dusting can occur. The thermodynamic assessments of Ref. 1 show that breakaway oxidation is not a credible threat in HTR He.

The above assessment will need to be checked when more precise compositional data become available from the PBMR and GT-MHR projects or a reference He composition is defined within the HTR-E project.

8 Outstanding materials issues

The HTR-M studies reviewed above have identified a number of materials issues that need to be addressed within the HTR-M project or elsewhere. A major factor in resourcing future HTR-M work is the existence of ‘cold’ and ‘warm’ design options for HTR reactor pressure vessels. It is not possible to predict which of these two approaches might be adopted in a future European HTR design so it is necessary to cover both options and their candidate material types i.e.

- C-Mn and Mn-Ni-Mo steels for cold vessels;
- Cr-Mo steels with 2 ¼ to 12% Cr for warm vessels.

Actions to tackle the identified issues are listed below in three categories.

a) Actions connected with the assembly and analysis of the HTR-M vessel database.
b) Actions connected with the HTR-M vessel experimental programme.
c) Actions requiring resources from outside the HTR-M project.

a) Actions connected with the assembly and analysis of the HTR-M vessel database.

This category includes comparisons between the design materials properties already provided in codes. Most design codes contain at least one material grade conforming to each of these material types. It is important to identify and understand any differences that arise at the grade level with respect to composition, manufacturing route and mechanical properties.

For cold vessels there is broad agreement between ASME and European codes as to the optimum composition, manufacturing route and properties of the prime candidate for the HTR RPV, Mn-Ni-Mo steel. Additional data are required to cover temperatures above the maximum of 370°C in LWR design rules. Mechanical properties data at temperatures above 370°C are available outside design codes and some sources were identified in Ref. 8. Data provided by HTR-M participants and from the open literature will be collected in the HTR-M database and analysed. Judgements about the adequacy of the properties database and about the effects of thermal ageing at higher temperatures will require information from outside HTR-M on the transients to be covered.

For warm vessels significant differences in design mechanical properties between various grades of 2¼ and modified 9Cr-1Mo steel are known to exist. The most important unresolved case identified in Ref. 8, concerns the creep strength of modified 9Cr-1Mo steel according to the ASME and RCC-MR design rules. For both types of material the HTR-M database will provide a resource covering ‘raw’ materials data, design materials data (from ASME, RCC-M, RCC-MR and other codes) and the basis on which the design data were derived from the raw data. Analysis of the database can then provide an understanding of the differences between codes and/or material grades with the possibility to identify new grades optimised for HTR vessel conditions. Actions relating to the properties of modified 9Cr-1Mo steel are also listed under the other two categories where requirements for new materials data have been identified.

b) Actions connected with the HTR-M vessel experimental programme.

It was decided early in the HTR-M programme to devote the experimental activities in the vessel area to modified 9Cr-1Mo steel. Subsequent studies have confirmed this decision.

The main point from this review concerns the relative effects of irradiation embrittlement and thermal embrittlement on toughness after exposure at 400-550°C. The HTR-M irradiation experiment has to cover the worst case of a heavy section weld. Irradiated and control specimens should therefore be exposed at the lowest credible beltline temperature and include weld metal specimens. As noted in Ref. 9 and Section 7 above thermal ageing effects in the simulated coarse-grained heat affected zones of relatively low residual C-Mn and Mn-Ni-Mo steels after exposure for a few hundred to a few thousand hours at 450-490°C were more severe than irradiation effects at 288°C. Equivalent exposure conditions can arise anywhere in warm vessels during either normal operation or transients so it is essential to obtain guidance on the potential severity of ageing in the HAZ of a modified 9Cr-1Mo weld. Subject to resource limitations and material availability it is therefore proposed that at least one additional series of control specimens be included in the HTR-M experimental programme to cover thermal ageing. Exposure conditions should be chosen on the basis of the best available estimates of GT-MHR vessel temperatures during normal operation and transients.

c) Actions involving resources outside the HTR-M Project.

The development of large ring forgings is beyond both the remit and resources of the HTR-M project. The requirement for such a development is independent of the choice between the cold or warm vessel options but the greatest difficulties are likely to arise in the case of a modified 9Cr-1Mo vessel. It is possible that a preliminary view might be obtained within one of the projects within the HTR cluster but the manufacture of trial forgings may only be feasible as a preliminary activity for a PBMR or GT-MHR construction project.

A second area within this category concerns long-term creep rupture testing covering products and temperatures relevant to a warm HTR vessel (400-500°C). High priority should be assigned to modified 9Cr-1Mo steel. The tests should cover base materials, weld metal and cross weld tests to check for Type IV cracking.

Engineering experiments with supporting materials tests may be required to establish design rules for the avoidance of ratchetting in cyclic softening materials such as modified 9Cr-1Mo steel. It is first necessary to establish (perhaps within the HTR-E project) whether ratchetting can pose a sufficiently severe threat to the integrity of HTR pressure vessels to require the use of relatively sophisticated design rules.

9 Conclusions

1. Different types of material are required to meet the requirements of ‘cold’ and ‘warm’ design options for HTR pressure vessels.
2. The ‘cold’ vessel design concept adopted for the PBMR project leads to the choice of Mn-Ni-Mo or C-Mn steel with grades similar to SA 508 Class 2 as prime candidates.
3. The ‘warm’ vessel concept adopted for the GT-MHR project leads to the choice of Cr-Mo steels with modified 9Cr-1Mo steel grades similar to ASME Grade 91 as prime candidates.
4. The ‘cold’ vessel concept allows advantage to be taken of PWR vessel technology and design rules. Additional rules and data may be required to cover transients involving temperatures exceeding the PWR design limit of 370°C.
5. High temperature design codes currently provide incomplete coverage of design rules and materials data for the main candidate warm vessel steel types, 2¼Cr-1Mo and modified 9Cr-1Mo. In many cases additional design materials data can be collected from other design codes or derived by compiling data from identified non-code documents.
6. The HTR-M vessel experimental programme will concentrate on the effect of irradiation on the strength and toughness of a heavy section weld in modified 9Cr-1Mo steel. It is recommended that the programme be extended to include the effects of thermal ageing on the coarse grained region of the weld heat affected zone.
7. Some actions required to provide HTR vessel manufacturing technology and design rules cannot be undertaken within the scope of the HTR-M project. These include:
   a) extension of PWR ring forging capabilities to the diameters and materials for cold and warm HTR vessels;
   b) long term creep tests at 400-500°C to resolve uncertainties concerning design stresses and weld factors for warm vessel materials;
   c) engineering assessments and perhaps experiments to establish ratcheting rules for warm vessels in cyclic softening materials such as modified 9Cr-1Mo steel.

10 References

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Table 1. HTR test reactors and designs for future plant

Country	Japan					South Africa	Russia + others	China	
System (1)	HTTR	MSF HTR	SSF HTR	HTR -GT 300	HTR -GT 600	PBMR	GT-MHR	HTR 10	MHGTR -IGT
Purpose (2)	Tests	H+E	E+DS	E	E	E	E	Tests	E+DS
Power, MW t	30	450-600	50	300	600	265	600	10	200
Core Type	Block	Block	Block	P. Bed	Block	P. Bed	Block	P. Bed	P. Bed
He Pressure, MPa	4	6	7	6	6	7	8	3	6
Core inlet, °C	395	350-550	490	550	460	536	490	300 (8)	550
Core Outlet, °C	850 950	850-950	850	900	850	900	850	900 (8)	900
RPV wall, °C	400	-	-	<350	460	300	440	-	<350?
RPV ID, m	5.5	-	-	7.27		6.2	7.3	2.5	6
Beltline thk., mm (3)(4)	122	-	-	150	-	140	200 approx.	100 approx .	-
Flange thk., mm (4)	300 approx	-	-	550 approx.	-	365	670	-	-
Vessel mass, te (5)	-	-	-	800 (V)	<1000 (V)	600	1362 (V+C)	142	-
Material	2¼ Cr 1Mo	Mod 9Cr	Mod 9Cr	Mn-Ni-Mo	Mod 9Cr	Mn-Ni-Mo	Mod 9Cr	C-Mn	-

Notes:

(1) Initials are as used by IEAE.
(2) E = electricity generation, H = hydrogen production, DS = de-salination
(3) BL = belt line, usually the minimum shell thickness.
(4) Thickness is called 'approx.' when estimated from a small-scale drawing.
(5) V = vessel only - excluding bolted closure. V+C includes bolted closure, vessel mass about 1000 te.

Table 2. HTR vessel candidate materials: chemical composition

	C	Si	Mn	P	S	Cr	Mo	Ni	V	Nb	N	Al	Cu
10Cr9MoVNb	0,08		0,30			8,6	0,6		0,15	0,10	?		
Russian grade	0,12	0,50	0,60	0,030	0,025	10	0,8	0,50	0,25	0,20			
Z10CDVNb9-1	0,08	0,20	0,30			8,0	0,85		0,18	0,06	0,030		
RCC-MR	0,12	0,50	0,50	0,020	0,010	9,0	1,05	0,20	0,25	0,10	0,070	0,040	0,10
Grade 91	0,08	0,20	0,30			8,0	0,85		0,18	0,06	0,030		
ASME/ASTM	0,12	0,50	0,60	0,025	0,025	9,5	1,05	0,40	0,25	0,10	0,070	0,040	
15Cr2NiMoV	0,12	0,17	0,30			2,20	0,50	0,80	0,08				
Russian grade	0,16	0,37	0,60	0,020	0,020	2,70	0,80	1,30	0,15				
2¼CrMo (7CD9-10)	0,07	0,15	0,30			2,00	0,90						
RCC-MR	0,11	0,35	0,70	0,020	0,015	2,50	1,10	0,30				0,020	0,20
10CD9-10		0,10	0,30			2,00	0,90						
AFNOR	0,15	0,50	0,60	0,030	0,035	2,50	1,10	0,30				0,025	0,25
Grade 22 CL 1			0,30			2,00	0,90						
ASME/ASTM	0,15	0,50	0,60	0,025	0,025	2,50	1,10						
16MND5		0,10	1,15				0,45	0,50					
RCC-M	0,20	0,30	1,55	0,012	0,012	0,25	0,55	0,80	0,01			0,04	0,20
A533 gr. B CL1		0,15	1,15				0,45	0,40					
ASME/ASTM	0,25	0,40	1,50	0,035	0,040		0,60	0,70					
A508 CL 2		0,15	0,50			0,25	0,55	0,50					
ASME/ASTM	0,27	0,40	1,00	0,025	0,025	0,45	0,70	1,00	0,05				
A42													
RCC-MR	0,18	0,25	0,60	0,030	0,008	0,25	0,07	0,30					0,30
A48			0,80										
RCC-MR	0,20	0,35	1,50	0,030	0,008	0,20	0,10	0,30	0,05				0,025
A516 gr. 60		0,15	0,85										
ASME/ASTM	0,27	0,40	1,20	0,035	0,040								
A516 gr. 70		0,15	0,85										
ASME/ASTM	0,31	0,40	1,20	0,035	0,040								

Notes:

AFNOR Z10CDVNb91 is X10CrMoVNb9-1 in EN designation

AFNOR 7CD9-10 is 7CrMo9-10 in EN designation

AFNOR 10CD9-10 is 10CrMo9-10 in EN designation

(Numbers 7 and 10 are selected to identify 2 grades differing in their specified carbon content)

AFNOR 16MND5 is 16MnNiMo5 in EN designation

Table 3. References giving design material data or a basis for their derivation for HTR vessel candidate materials within the temperature limits shown.

10Cr9MoVNb	tensile	(13)	→ **600 °C**
	creep	(14)	→ **600 °C**
Z10CDVNb9-1	tensile	(16) - Appendix A3-18S) + (15)	→ **600 °C**
	creep	(18)	→ **650 °C**
grade 91	tensile	(15) or (21) → **538 °C**	(21) → **649 °C**
	creep	(21)	→ **649 °C**
15Cr2NiMoV	tensile	(12) → **350 °C**	
	creep	(?)	
7CD9-10	tensile	(16 – Appendix A3-16S)	→ **550 °C**
	creep	(16 – Appendix A3-16S)	→ **550 °C**
10CD910	tensile	(16 – Appendix A3-14S)	→ **550 °C**
	creep	(16– Appendix A3-14S)	→ **550 °C**
grade 22	tensile	(15)Section II – D) → **482 °C**	(15 Subsection NH) → **649 °C**
	creep	(15 Subsection NH)	→ **649 °C**
16MND5	tensile	(17) → **370 °C**	(23)→ **600 °C**
	creep	(22)	→ **600 °C**
A533 grade B CL 1	tensile	(15 Section II – D) → **371 °C**	(22)→ **600 °C**
	creep	(22)	→ **600 °C**
A508 CL 2	tensile	(15 Section II – D) → **371 °C**	
	creep	no data	
A42	tensile	(16 – Appendix A3-11S) → **350 °C**	(23) → **600 °C**
	creep	(21)	→ **500 °C**
A48	tensile	(16 – Appendix A3-12S) → **350 °C**	(23) → **600 °C**
	creep	(23)	→ **500 °C**
A516 gr. 60	tensile	(15 Section II – D) → **371 °C**	(24) → **538 °C**
	creep	(24)	→ **566 °C**
A516 gr. 70	tensile	(15 Section II – D) → **371 °C**	(24) → **538 °C**
	creep	(24)	→ **566 °C**

Table 4. Recommended values of creep stress to rupture factor J_r for modified 9Cr-1Mo welds.

Temperature °F	900	1000	1100	1200
Temperature °C	482	538	593	649
J_r	0.93	0.90	0.85	0.76

Figure 1. Effect of irradiation on the Ductile to brittle transition temperatures of pressure vessel steels

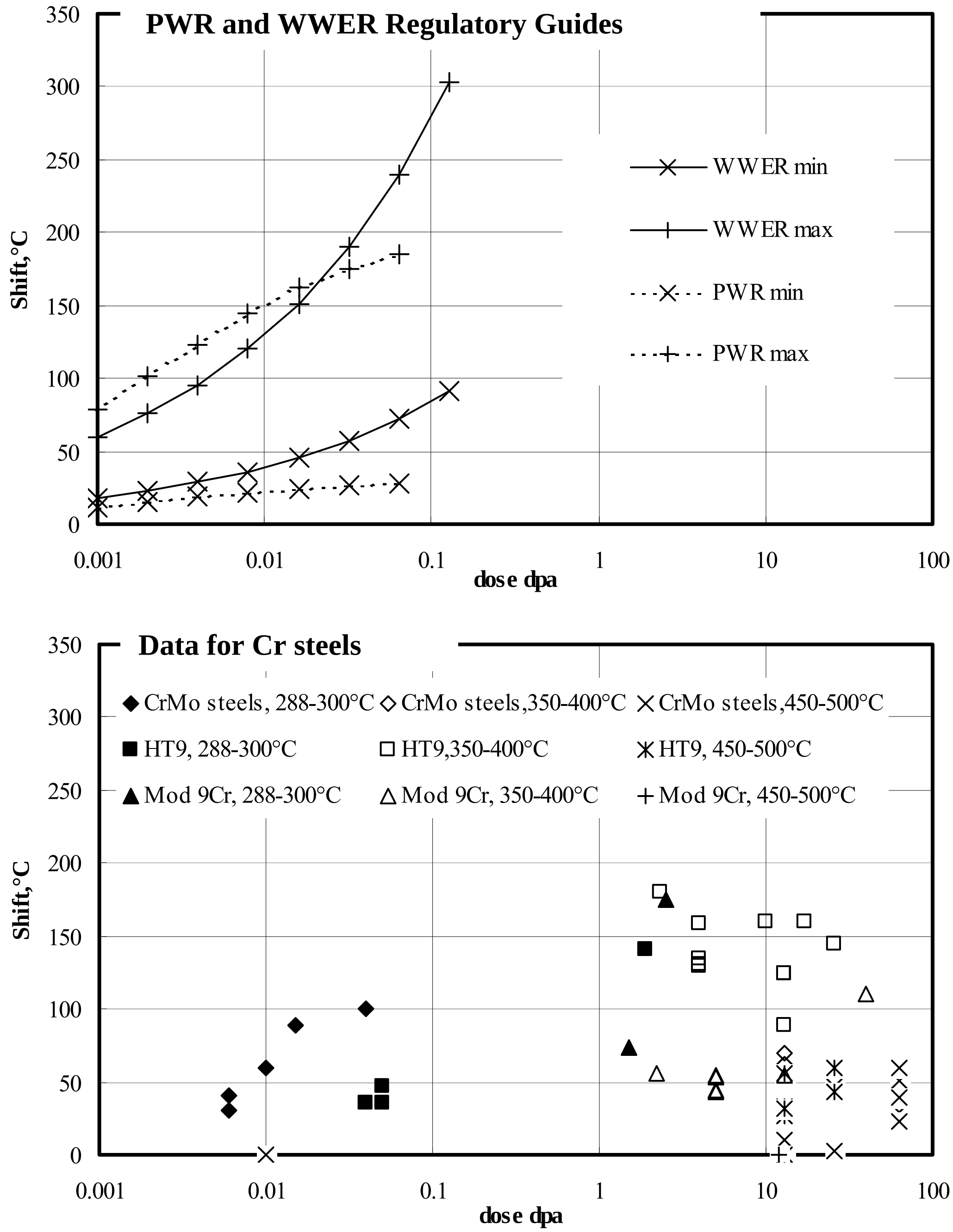
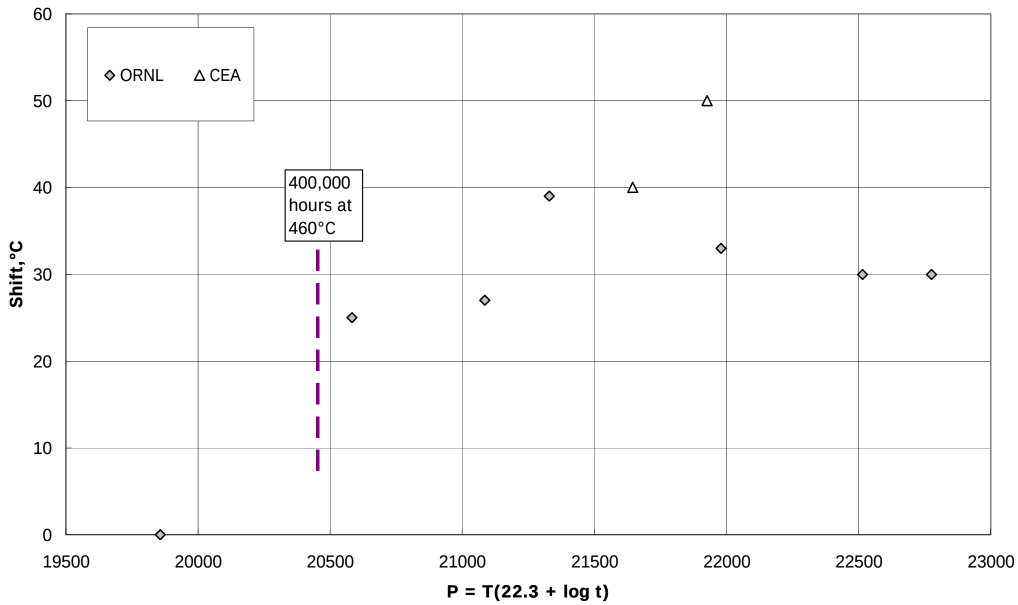


Figure 2. Effect of thermal ageing on the toughness of modified 9Cr-1Mo steel base material

a) Ductile-Brittle Transition Temperature



b) Upper Shelf Energy