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

HTR-M

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

CONTRACT N°
FIKI-CT-2000-00032

HTR-M Project:- Preliminary Review of RPV materials used on HTR and other gas cooled reactors.

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UK

Dissemination level : CO
Document Number: HTR-M-01/9-D-1.1.9

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C6463/TR/004
Issue 02
June 2004

DOCUMENT ISSUE RECORD
(engineering documents)

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Document Title :	HTR-M Project:- Preliminary Review of RPV materials used on HTR and other gas cooled reactors.
Project Reference:	C6463

Purpose of Issue : Final Issue

Security Class : Commercial-in-Confidence

Issue	Description of Amendment	Originator/ Author	Checker	Approver	Date
01	First Issue				
		R. D. W.Bestwick	A. A. Braithwaite	D. E. Buckthorpe	September 2001
02	Final Issue including comments from HTR-M partners	R. D. W.Bestwick	A. A. Braithwaite	D. E. Buckthorpe	June 2004

Total number of pages:	Intro:	(i)-(v)	Text	1-5	Tables	T1-T2	Figures	F1-F8	Appendices	

Previous issues of this document shall be destroyed or marked **SUPERSEDED**

Distribution: S. Casalta (EU), HTR-M Partners via HTR-M Forum on SINTER web site

NNC : D. Buckthorpe, A. A. Braithewaite, EDMS

3050aJan01 Controlling procedure - QP11, QP40 *Docprodref*

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Summary

The HTR-M project has as one of its objectives the improvement of the reactor pressure vessel (and other pressure vessels used to contain primary circuit components) for future HTRs. This document provides a review of currently used materials as a first step towards identifying a material or materials for study during the welding, irradiation and mechanical testing phases of the HTR-M project. The review is also aimed at identifying items to be raised in discussions between HTR-C and HTR specialists in China and Japan.

1. Introduction

The HTR-M project has as one of its objectives the improvement of the reactor pressure vessel (and other pressure vessels used to contain primary circuit components) for future HTRs. 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 property data. This review is a first step towards identifying a material or materials for study during the welding, irradiation and mechanical testing phases of the HTR-M project. The review is also aimed at identifying items to be raised in discussions between HTR-C and HTR specialists in China and Japan.

2. Pressure Vessel Designs

Until the late 1980's the HTR was considered alongside other systems for large-scale electricity production. Reactors with power outputs up to 2 GW (th) were under consideration based on the use of pre-stressed concrete pressure vessels.

In the 1980's it was realised that smaller HTRs with inherently safe characteristics offered potentially significant advantages with respect to simplicity and public acceptance. One characteristic of these systems is a steel pressure vessel which can transmit stored and decay heat to its surroundings at a sufficient rate to maintain acceptable temperatures within the vessel including peak core temperatures. Modular systems with gas turbines at around 350 MW (th) are now judged to be economically attractive due to the elimination of complex safety systems. Other potential applications include:

- Combined electricity generation and process heat for desalination plants.
- High temperature process heat for hydrogen and synthetic fuel production.

Design and R & D activities have been undertaken in a number of countries. The IAEA International Working Group on Gas Cooled Reactors (IWG-GCR) has provided a valuable forum for the exchange of information and opinion. Much of the information considered in this review has been extracted from a document edited and published by IAEA on the basis of contributions from China, France, Germany, Indonesia, Japan, the Netherlands, Russia, South Africa, Switzerland, the United Kingdom and the United States of America (Ref. 1). Japan has proved to be a particularly fruitful source of forward-looking plant design concepts. A Japan Atomic Energy Research Institute (JAERI) document (Ref. 2) gives the status of several plant designs as of early 2000. Table 1 lists test reactors and plant designs described in Refs. 1 and 2 along with data relevant to the choice of material.

3. Materials used in present and future plants

Table 1 lists test reactors in Japan (HTTR) and China (HTR 10) along with designs under consideration in Japan, China, Russia and South Africa. It should be noted that the design pressures and internal diameters of the HTR pressure vessels fall within relatively narrow ranges. This is due to the constraints imposed by requirements for passive heat removal under loss of circulation or loss of coolant scenarios and the allowable stress levels in various design codes. Figure 1 shows the allowable primary membrane stress limit S_m or S_{mt} as a function of temperature for the four materials listed in Table 1. S_m

is the time independent stress limit, determined from the tensile properties. S_{mt} is the corresponding time dependent stress limit obtained from both tensile properties and creep properties.

Two types of material are 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.

In China silicon killed C-Mn steel according to ASME SA 516-70 has been specified for both the HTR 10 test reactor and a future power reactor. This material 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. Long term operating and/or design temperatures are limited to 370°C. Similar limits apply to the SA 508 forgings used widely in LWR pressure vessels and proposed for pebble bed reactors of approximately 300MW_{th} in both South Africa (PBMR) and Japan (HTR-GT 300). It should be noted that the SA 508 specification covers a number of grades, some of which are not suitable for pressure vessels. In terms of the current designations in ASME specifications 'SA 508 type' material is taken hereafter to mean SA 508 Grade 3 Class 1 or the equivalent material in plate form SA 533 Type B Class 1. Equivalent European designations are 20MnMoNi 5 5 (Germany) and 15MND 5 or 18MND 5(France). Except for HTR-10 the above systems have core inlet gas temperatures close to 550°C so the integrity of the RPV depends on the performance of insulation in the form of fibre blanket and/or stagnant layers of He. Figures 2 and 3 show the operating and allowable levels of membrane stress in the beltline of each RPV where the minimum material thickness occurs. It is clear that levels of primary membrane stress have been set on a conservative basis when compared with a large (1100 MW) PWR vessel such as Sizwell B.

In both the HTTR test reactor and larger power reactors of 450-600 MW_{th} the RPV temperature is close to the core inlet temperature due to the use of inlet (or intermediate) temperature helium to cool the inside surface of the vessel. Vessel temperatures are in the range 395-460°C under normal operating conditions.. Figure 4 shows that the HTTR inlet temperature of 400°C permits the use of 2¼ Cr-1Mo steel with a margin of approximately 25% on allowable stress and about 40°C on temperature. Figure 4 also shows the situation if the simpler design rules of ASME VIII Division 1 are used to set the allowable level of membrane stress intensity. The margin on stress intensity at 400°C is reduced because a higher factor is applied to the tensile properties but the temperature margin is apparently increased because ASME VIII Division 1 bases allowable stresses on creep strength at 10^5 hours rather than $3x10^5$ hours. It should be noted that additional restrictions are incorporated in the design by rule approach of ASME VIII Division 2. ASME III-NH (formerly Code Case N47) uses the ASME VIII Division 1 formulation for design conditions and the S_{mt} limit for normal operating conditions. The RCC-MR rules use only the S_{mt} limit.

Modified 9Cr 1Mo steel has been specified for both large and small reactors with the beltline region of the vessel running at the core inlet temperature. This material 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 were extended during the European Fast Reactor (EFR) project to cover its use for tubeplates, shells and hemispherical ends in steam generators with outlet steam temperatures near 500°C. The EFR values of S_{mt} are shown as a function of temperature in Figure 5 along with approximate primary stress intensity values for the GT-MHR vessel. The operating point corresponds remarkably closely to the allowable stress given that the beltline thickness is an estimate scaled from a small-scale reproduction of a drawing. It should also be noted that Russian design rules use different factors in deriving design stresses from specified mechanical properties.

4. Compatibility considerations

The above rationale for materials selection in terms of design stresses does not take account of any additional material thickness required to allow for the effects of the He environment.

HTR gas chemistry is dominated by the impurities H_2, H_2O, CO, CO_2. 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. Three factors will therefore control the He impurity levels:

- The hydrogen and oxygen supply 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.

Table 2 gives information on He impurity levels from various sources. Thermodynamic assessments have been made for these atmospheres (Ref. 10). The results of the assessments are presented in terms of the carbon activity and oxygen potential (notional partial pressure of oxygen) on the phase diagrams shown in Figures 6-8. Points are included for Cases 2, 3, 5, 10, and 17 from Table 2 and a typical AGR gas composition with $4x10^6$ Pa CO_2, $4x10^4$ Pa CO, $1.7x10^4$ Pa H_2O and 800 Pa H_2.

Figure 6 shows the results at temperatures relevant to PBMR. A specification for the PBMR gas composition has not been found but it can be noted that:

- All of the HTR gas compositions and the AGR gas have carbon activities greater than unity at 300 and 350°C i.e they will tend to deposit carbon.
- The five selected He compositions 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.
- The AGR gas has a high carbon activity and oxygen potential. These gas characteristics have the potential to lead to breakaway oxidation. The low Si content of vacuum-carbon deoxidised pressure vessel steels would be a disadvantage at AGR levels of oxidation potential. These conditions can be reached in He only by increasing the water content by about 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.

Figures 7 and 8 show the results at temperatures relevant to GT-MHR. It should be noted that the GT-MHR composition is an upper bound to both the range of compositions allowed in the plant and the selected He compositions from Table 2. The selected He compositions are 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 is therefore decarburisation, particularly at 540°C. By contrast Cr and its carbides will be oxidised in all of the selected 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 metal losses due to the He coolant allowances for coolant will not be due to the formation of oxide scales but to the effects of carburisation in cold wall vessels or 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. It has already been shown that breakaway oxidation is not a credible threat in HTR He. The possibility that long term carburisation of cold wall material might lead to metal dusting cannot be excluded on thermodynamic grounds. In this case kinetic data are required to evaluate the threat to vessel integrity. It can be noted that two of the selected He compositions are essentially neutral with respect to the equilibrium between Fe_3C and Fe_3O_4 . These compositions are very different in terms of the total impurity levels 5.5 Pa for the Direct cycle case and 91 Pa for GT-MHR. If the metal dusting problem is judged to be a threat at cold wall temperatures then an alternative approach based on control maintaining equilibrium values of oxidation potential could be adopted. It seems therefore that metal coolant interactions can be allowed for in design by specifying allowances for metal losses that are a small proportion of the minimum beltline thickness from basic design calculations.

5. Implications for HTR-M Reference Material

From the above survey it can be seen that most designs of future plant make use of the either SA 508 type steel for ‘cold wall’ 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 long term design strength is twice that of 2¼ Cr-1Mo steel.

Existing LWR pressure vessels in SA 508 type material and its European derivatives encompass the conditions in proposed cold wall 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 about 370°C.

A substantial data base 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 (2 ¼ Cr-1Mo grades). There should be relatively few problems in assembling an adequate data base extending to temperatures involved during transients.

Except for the possibility of carburisation at cold wall vessel temperatures, coolant compatibility issues are not expected to have a strong direct impact on materials selection because metal loss allowances will be a small proportion of the pressure vessel wall thickness.

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. Since this is also the most frequently occurring material in Table 1, it can be proposed as the reference material for the fabrication and mechanical testing programme of HTR-M. Future compatibility studies should cover both cold wall and a warm wall materials and temperatures.

6. 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	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 appro	-	-	550 approx	-	365	670	-	-
Vessel mass, te (5)	-	-	-	800 (V)	<1000 (V)	600	1362 (V+C)	142	-
Material	2¼ Cr	Mod 9Cr	Mod 9Cr	SA508	Mod 9Cr	SA508	Mod 9Cr	SA516 -70	-

Notes:

(1) Initials are as used in Refs. 1 and 2.

(2) E = electricity generation, H = hydrogen production, DS = de-salination

(3) BL = belt line, usually the minimum shell thickness. Max usually refers to that part of the cylinder containing the ‘cross vessel’ connection between the RPV and the power system vessel.

(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. He impurity levels

Case(s)	Description	Pressures in Pa				
		H_2	CO	H_2O	CO_2	CH_4
1 & 2 (Ref. 3)	Dragon Normal Range	1 1.5	0.5 0.8	0.01 0.015	0.005 0.08	0.1 0.15
3 (Ref. 3)	Realistic Direct Cycle	3	2	0.5	-	-
4 (Ref. 3)	CIIR Early Tests	25	25	5	-	-
5 (Ref. 4)	Oldbury B standard	20	0.5	0.5	-	2
6 (Ref. 4)	Oldbury B boiler leak	50	50	5	-	5
7 & 8 (Ref. 5)	ORNL test programme	40 40	1 12	0.01 1	0.02 0.02	0.07 0.8
9 (Ref. 6)	PNP He	50	1.5	0.15	-	2
10 (Ref. 7)	He-1 (JAERI Type B)	20	10	0.1	0.2	0.5
11 (Ref. 7)	He-2	5	2.5	0.1	0.2	0.5
12 (Ref. 7)	He-3	50	0.3	0.1	-	0.5
13 (Ref. 7)	He-4	50	0.3	0.005	-	0.5
14 & 15 (Ref. 8)	JAERI Type B Range	20 21	10 11	0.08 0.12	0.2 0.3	0.5 0.6
16 (Ref. 9)	PNP	50	6.5	0.1	-	2
17 (Ref. 1)	GT-MHR normal	<35	<42(1)	<14	(1)	-

(1) Ref. 1 gives an upper limit of 42 Pa for CO + CO_2 . The calculations described in the Appendix were based on a CO/CO_2 ratio of 50:1.

Figure 1. Allowable stresses for RPV materials

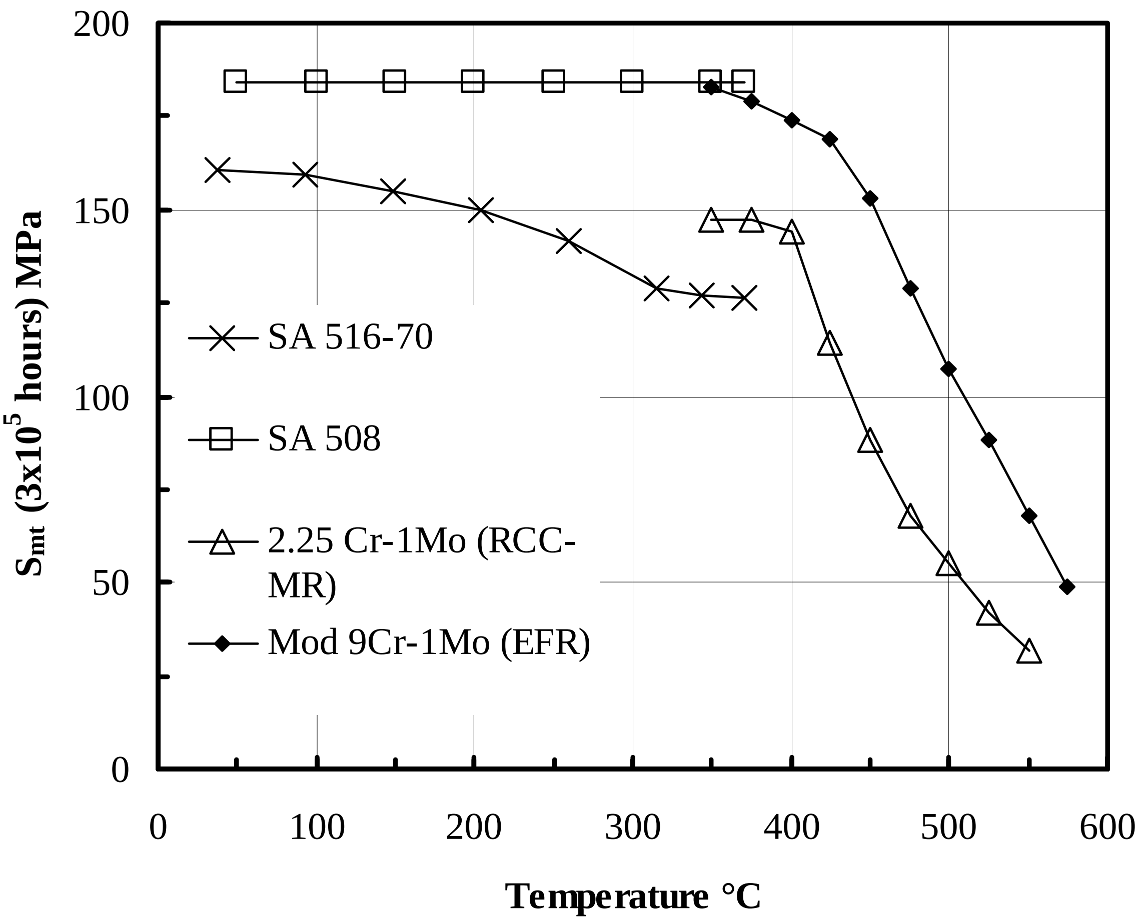


Figure 2. Membrane stress intensities and allowable stresses for SA 516-70 steel used in HTR-10

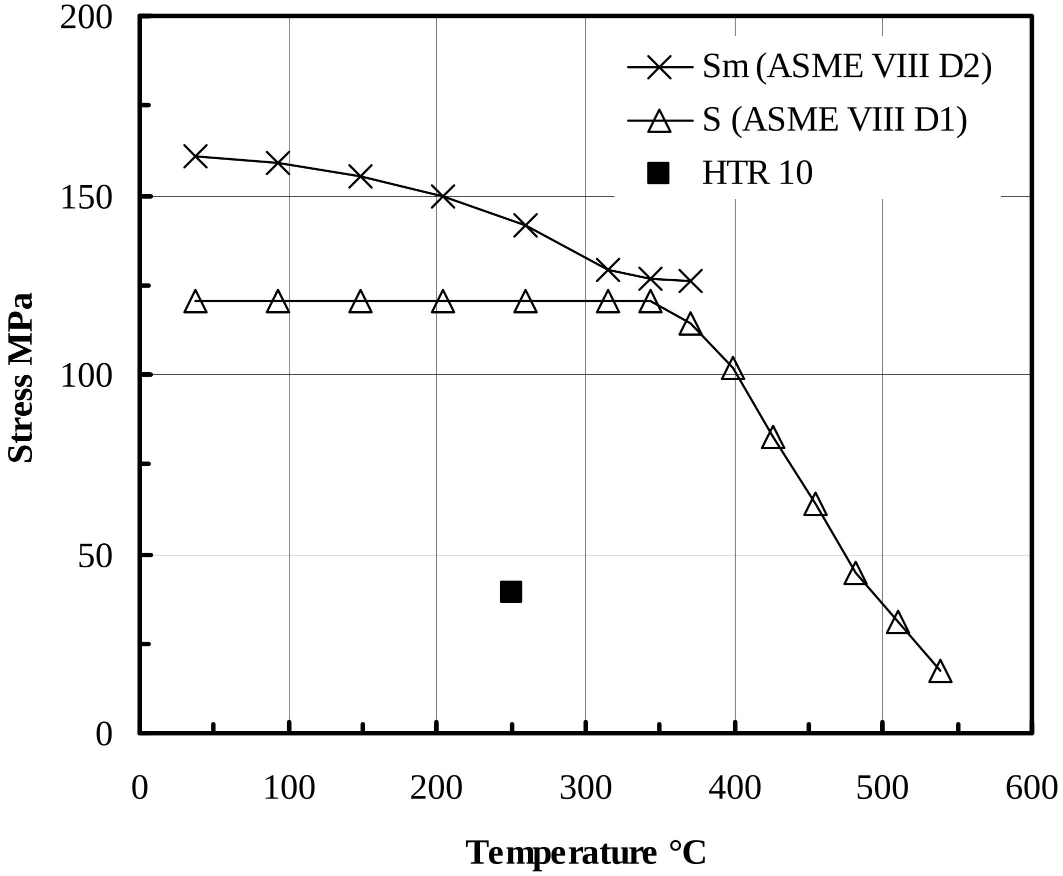


Figure 3. Membrane stress intensities and allowable stresses for SA 533B and similar LWR steels

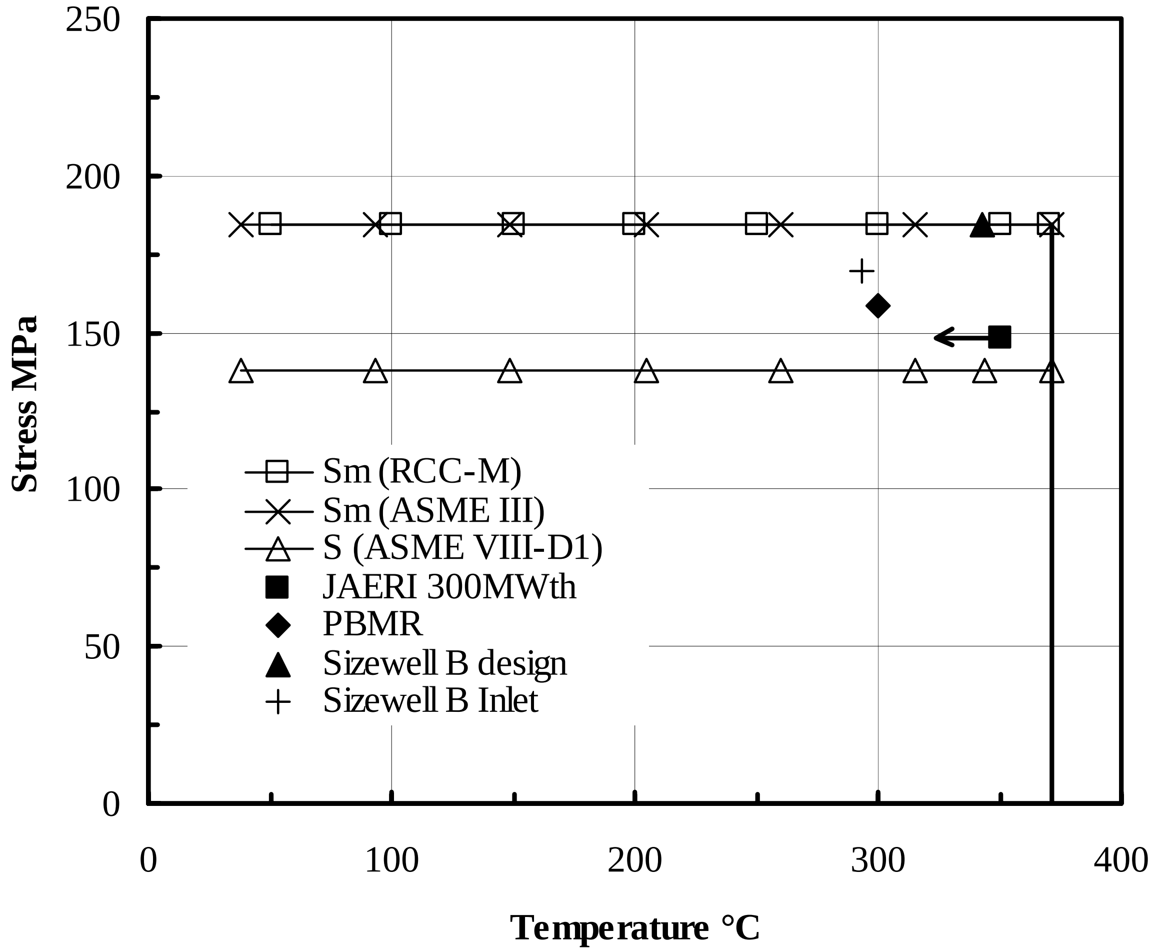
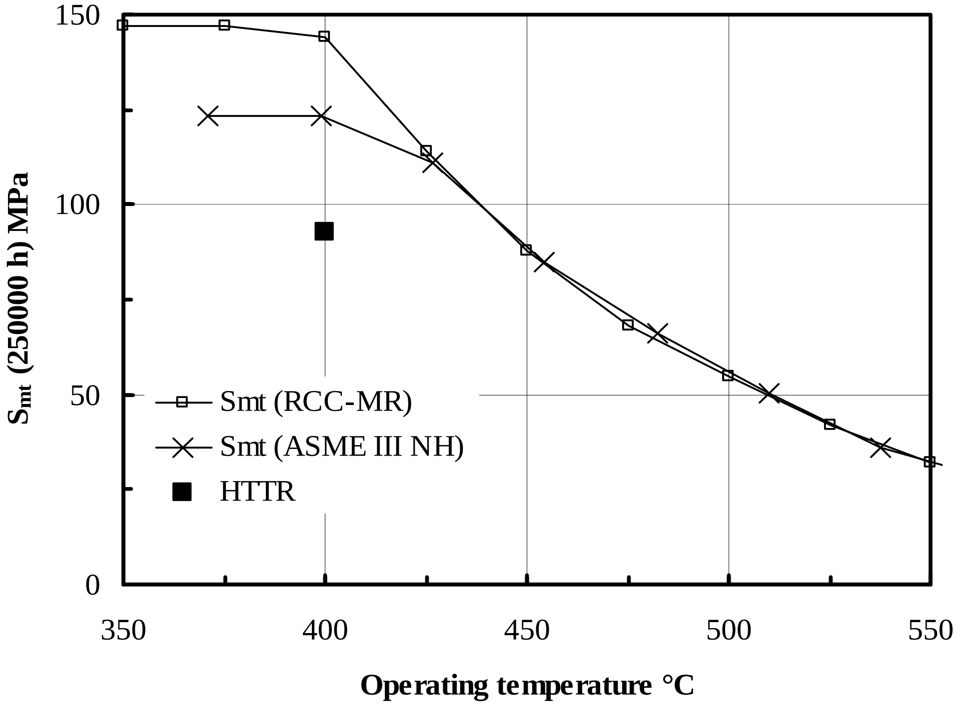


Figure 4. Membrane stress intensity and allowable stresses for 2¼Cr-1Mo steel



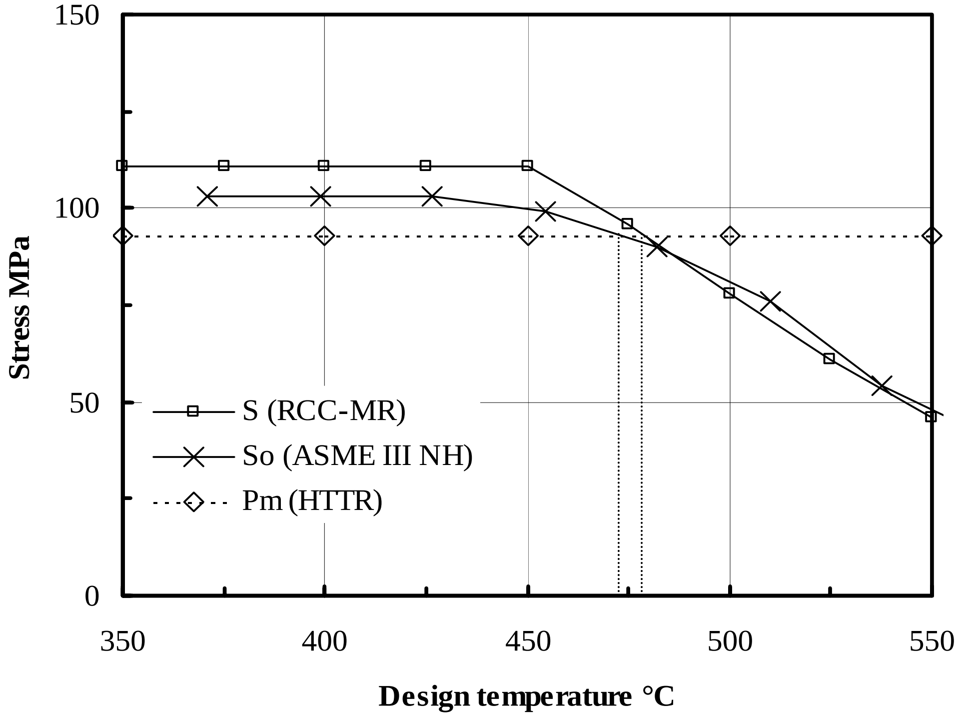


Figure 5. Membrane stress intensity and allowable stresses for Mod 9Cr-1Mo steel

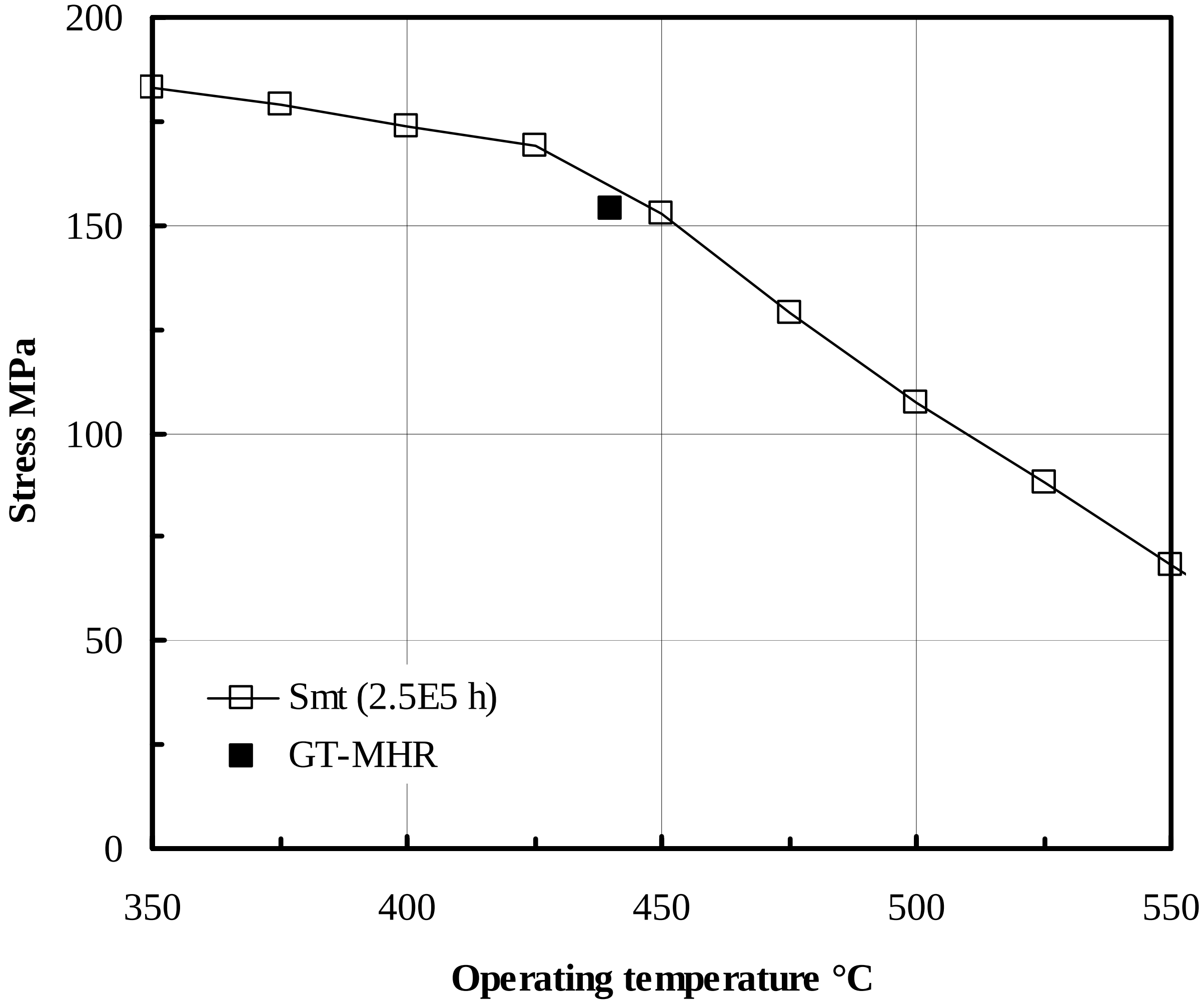
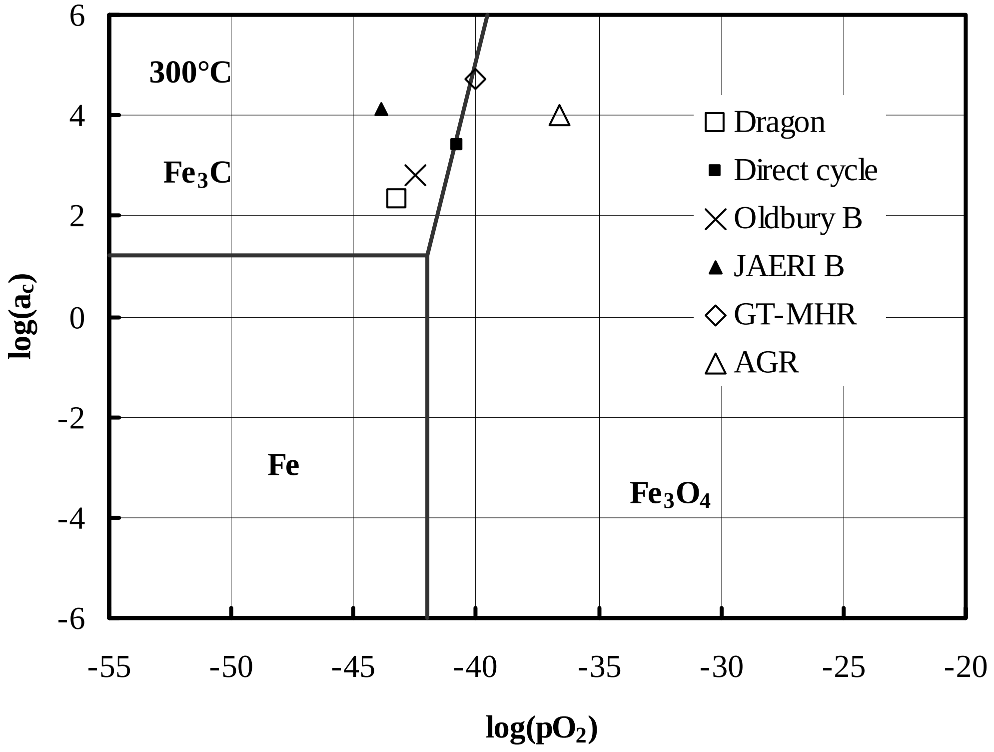


Figure 6. Phase diagrams for Iron at 300 and 350°C



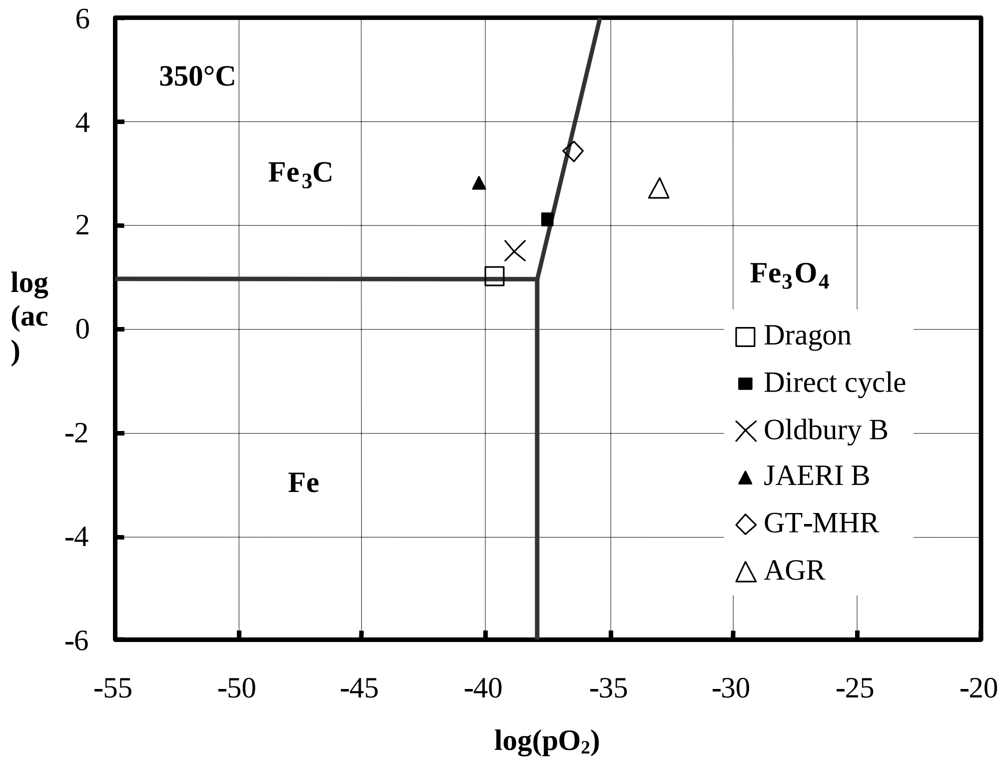
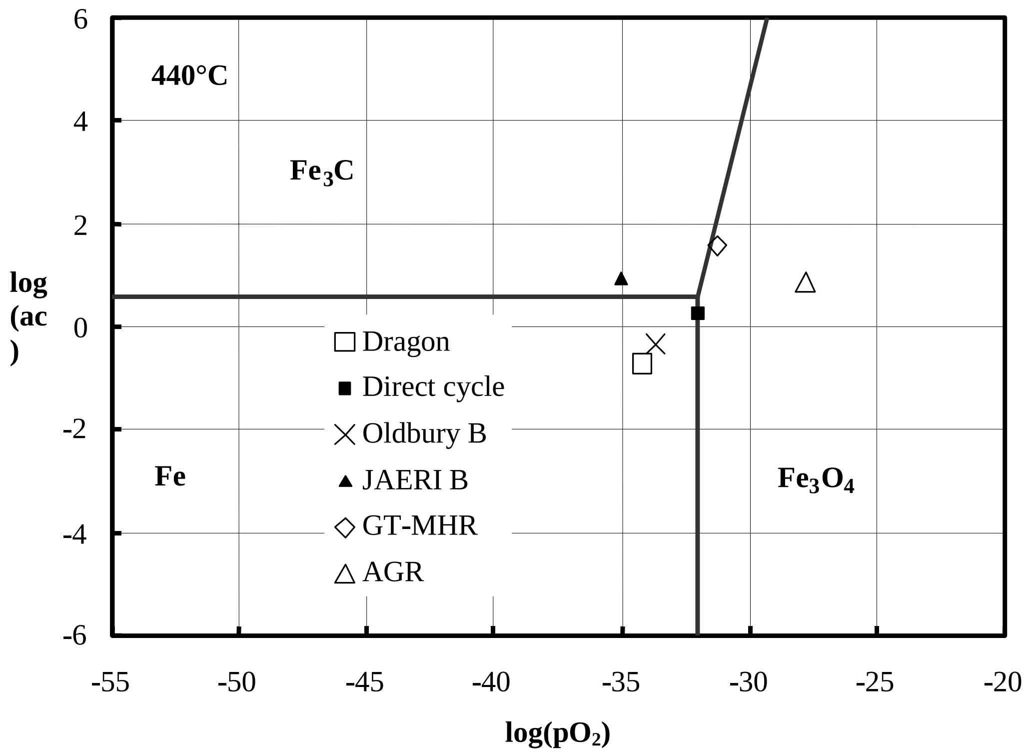


Figure 7. Phase diagrams for Iron at 440 and 540°C



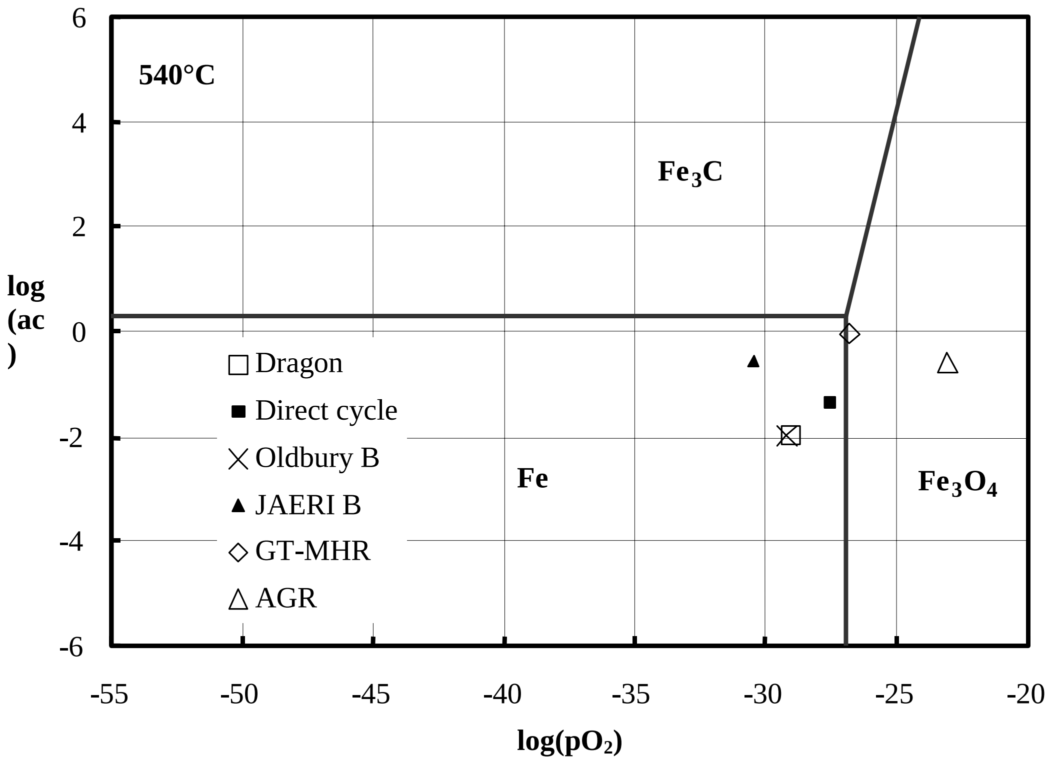


Figure 8. Phase diagrams for Chromium at 440 and 540°C