

CANDIDATE MATERIALS FOR ADVANCED HTGRS

Lineo Makhele-Lekala¹

Research Centre Juelich, IWV-2
52425 Juelich, Germany
Phone: +49 2461-615096
Fax: +49 2461-616464
E-mail: l.makhele@fz-juelich.de

Philip Ennis

Research Centre Juelich, IWV-2
52425 Juelich, Germany
Phone: +49 2461-614132
Fax: +49 2461-613699
E-mail: p.j.ennis@fz-juelich.de

Florian Schubert

Research Centre Juelich, IWV-2
52425 Juelich; Germany
Phone: +49 2461-613229, Fax: +49 2461-613699
E-mail: f.schubert@fz-juelich.de

ABSTRACT

The performance of candidate alloys for High Temperature Gas-cooled Reactors (HTGR) is reviewed and past experiences from the German HTR materials programmes are discussed. The components under consideration are the pressure vessel, hot ducting, intermediate heat exchangers (IHX) and turbines. Results concerning the deformation behaviour of the martensitic 9%Cr-steel (P92), a candidate material for the pressure vessel, under simulated accident conditions are presented. The results of the tests on P92, in which the specimens were heated from room temperature at a rate of 600 °C/h under constant load, showed that P92 could withstand temperatures of up to 700°C at a load of 100 MPa before catastrophic deformation. Wrought high temperature alloys are required for the hot gas ducts and heat exchanger tubing. Alloy 617 (for temperatures up to 950 °C) and Alloy 800H (for temperatures up to 850 °C) have been well characterized in the German programmes. The reactions between the metal and the impurities in the helium coolant can lead to carburisation or decarburisation, depending on the gas kinetics. Carburisation reduces low temperature ductility and decarburisation leads to a severe loss in creep strength, but control of the impurity contents can keep these effects within acceptable limits. For large-scale rotors or discs, there are alloys available for temperatures up to 600 °C. For the turbine blades, cast Ni-base alloys have the highest rupture strengths and there is extensive experience with alloys that can operate at temperatures up to 900 °C.

Keywords: pressure vessels, hot ducting, heat exchangers, turbine materials

1. INTRODUCTION

Historically, the development of the high temperature gas cooled reactor HTGR began in the 1950s, with significant advances in system design and fuel concepts during the 1960s and 1970s. The designs may be divided into two main groups, namely, steam-cycle HTGRs and modular gas turbine HTGRs. In all HTGR designs, helium

¹ Permanent Address: lineo.lekala@pbmr.co.za

serves as the primary coolant. The early HTGR designs (Rickard and Fortescue 1982) included, amongst others, the Dragon Reactor, Arbeitsgemeinschaft Versuchsreaktor (AVR), and Peach Bottom, which all utilized steel primary system vessels. Furthermore, the Dragon reactor incorporated spherical graphite fuel elements containing coated particles of highly enriched uranium carbide (Gulden and Nickel 1977) with the core exit and inlet helium temperatures of 750 and 350 °C, respectively. On the other hand, the German 15 MWe AVR, which is a pebble-bed type HTGR and began operation in 1967, operated with a core outlet temperature as high as 950°C (Marnet et al 1982, Kröger et al 1988). It had a steel containment vessel and used coated particle fuel embedded in graphite spheres 6 cm in diameter. The Peach Bottom HTGR (40 MWe) in the US operated without fuel failures for 897 equivalent full power days with fuel particles that were coated with isotropic pyrolytic carbon. The subsequent prototype HTGR plants, such as the Fort St. Vrain (FSV) and the Thorium High Temperature Reactor (THTR-300) had the primary systems enclosed in pre-stressed concrete reactor vessels (PCRVs). The FSV reactor was designed with a core of hexagonal, graphite-block fuel elements and reflectors with fuel in the form of ceramic-coated (TRISO) particles, once-through steam generator modules producing 538 °C superheated steam, and steam-turbine-driven axial helium circulators. The THTR-300 reactor, which was completed in 1984, demonstrated the desired safety features characteristics of the pebble bed, as well as good fission product retention of the fuel elements, before it was shut down permanently in 1991, on socio-political grounds. The aforementioned HTGR designs are all based on steam plants for power generation.

In recent times there has been a renewed interest in studies involving High Temperature Gas-cooled Reactors (HTGRs), for example, the HTR-M & HTR-M1 projects within the 5th and 6th Framework Programmes of the European Union, and the PBMR project in South Africa. The reason for this emergence of interest is due, in part, to the high energy demands in the world and the new developments in high temperature resistant materials. The latter point is crucial since the possible maximum temperature achievable in the designs of safe reactors will be limited by the availability of suitable constructional materials.

One of the most important factors in the development of modern industrialised societies is the reliability of energy supply at reasonable cost. However, societies are becoming increasingly concerned about the environmental impact of the energy production (mainly the emission of greenhouse gases, such as CO₂) and the need to conserve energy resources for the future generations. Although new technologies such as fuel cells and the use of renewable energy have emerged, the conversion of fossil fuels will continue to make a significant contribution to power generation in the next decades. The by-product of the conversion of fossil fuel will remain CO₂ emission. Nuclear power plants do not emit CO₂, hence the renewed global interest in their development, especially in the European Community, the USA, South Africa, China and Japan. In the 1970s and 80s HTGR concept proved its viability via the construction and operation of experimental and prototype plants in USA, Great Britain, Japan and Germany, with helium temperatures up to 950 °C. The availability of the high temperature resistant structural materials limits the helium temperature to around 1000°C. Although there has been an enormous development in the past decades of materials for highly loaded components in fossil-fired steam and gas turbine, it must be discussed how these developments may be applicable for the HTGR vision.

In order to design safe reactor plants, materials that are capable to withstand extreme service exposures conditions are required. The components of interest are pressure vessel materials, materials for helium turbines and materials for IHX and hot ducting. For the above components, several key technologies such as the behaviour of structural materials under the highest constraints in helium atmosphere must be mastered.

2. MATERIALS FOR THE REACTOR PRESSURE VESSEL (RPV)

The 9% Cr steels are favoured candidates materials for application as RPVs in future reactors. In the Very High Temperature Reactors (VHTR), where the helium pressure is 7 MPa, the nominal operating temperature of the vessel is expected to be about 500 °C under normal operational conditions and 650 °C during 50 h accident condition (Billot, 2003). For the GT-MHR, the operating temperatures are around 400 °C to 460 °C with potential temperature excursion of up to 570 °C and the prime candidate material is the modified 9Cr1Mo steel similar to the ASME Grade 91 (Buckthorpe, 2003). The current PBMR design uses the SA 508 Grade 3 with 300°C as the normal operating temperature and 430 °C under design basis events, while for the future planned VHT-PBMR whose RPV normal operating temperature would be 300 °C and the design basis events as 470°C has the ASME 9Cr1Mo as the candidate materials (Fazluddin, 2004).

The pressure vessels material of the German modular HTR design is 20MnMoNi 5 5 grade which is similar to the US material (ASME) SA 508 Grade 3 and the French material (RCC-MR) A3-185. In Germany, this material is approved for operation at temperatures below 350 °C. This means the vessel must operate in the negligible creep regime, so that the design is made with time-independent strength values (S_m). Figure 1 shows the temperature regimes in which time-independent (S_m) strength values may be used, for several candidate materials.

In the German design, the low operating temperature could be achieved by using a cooled structure for the outer wall of the pressure vessel. Furthermore, a limitation of the allowable accumulated neutron irradiation (e.g. 10^{19} n/m² fast neutrons) by different methods of shielding is required.

The advantages of the 20MnMoNi 5 5 type steels are (Forch. K. 1980):

- Manufacturing procedure and experiences exist for forging large diameter, (e.g. thick rings)
- For this material a through-hardening can be assured (i.e. the heat treatment is controlled).
- Welding procedures have been evaluated for nuclear power plant components.

If the material temperature exceeds the given maximum allowable material temperature of 370 °C, for example during emergency conditions, the deformation under creep becomes more important, and the deformation behaviour is now time dependent, which involves the risk of permanent deformation, and a loss of component integrity and the function.

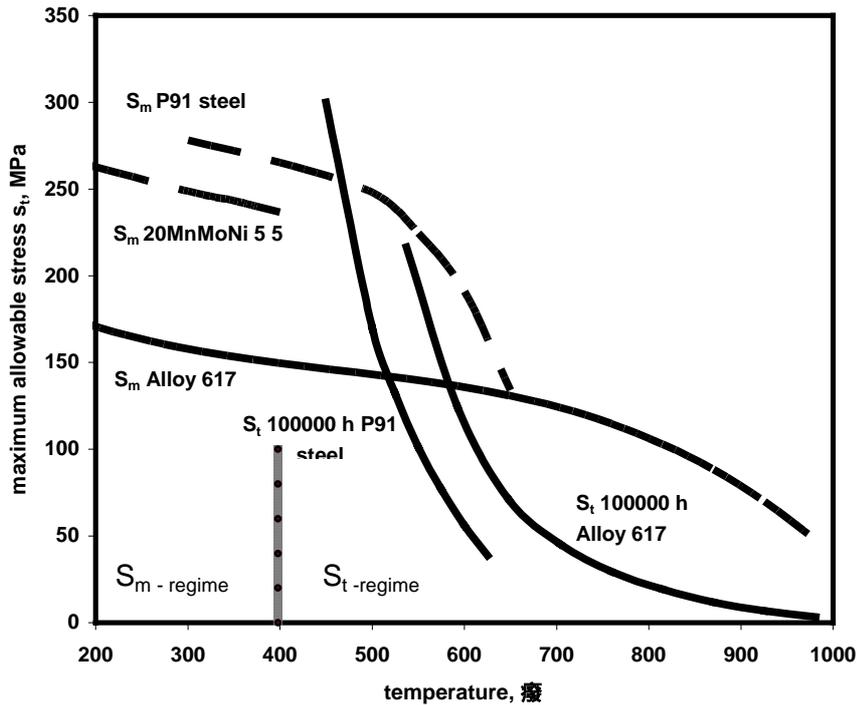


Figure 1 Temperature independent (S_m) and temperature dependent (S_t) design regimes for 20MnMoNi 5 5, P91 steel and Alloy 617

With regard to the 9% Cr steels, there has been an intensive research for application in steam temperatures above 550°C. The improved 9% Cr steels (P91, E911 and P92) offer substantially increased stress rupture than the low alloyed RPV steels and therefore should provide larger safety margins in the design and decrease the need for any active cooling of the pressure vessel. Figure 2 compares the stress rupture strengths of power station steels on the basis of the temperature at which the 100 000 hours stress rupture strength is 100 MPa.

In modern fossil-fuelled power plants, operating at temperatures up to 600 °C, modified 9%Cr steels are widely used. This temperature is higher than the operating temperatures of the pressure vessel materials proposed for the nowadays HTGRs. The stress rupture strength advantage of P92 compared with the other high temperature Cr steels is shown in Figure 3. As far as the RVPs are concerned, however, there remain some unsolved problems for the 9 % Cr steels. The steels rely on tempered martensite and fine carbide precipitates to produce the stable, creep resistant sub-structure for long-term service. At slow cooling rates, as could be the case for thick-walled components, the martensitic transformation could be suppressed and the creep strength is then lost (Ennis et al 2001). However, for P92, the TTT diagram (Figure 4) shows that cooling from 800 to 500 °C in less than about 5 hours will result in a martensitic microstructure which can be tempered in any way. The 9% Cr steels are also susceptible to creep failure in the heat-affected zone. Investigation of these two phenomena is required if the 9% Cr steels are to be used for the RPV to take advantage of higher strength under upset conditions. In addition, there is

more information concerning the effect of irradiation and the helium environment on the mechanical behaviour of 9%Cr steels required.

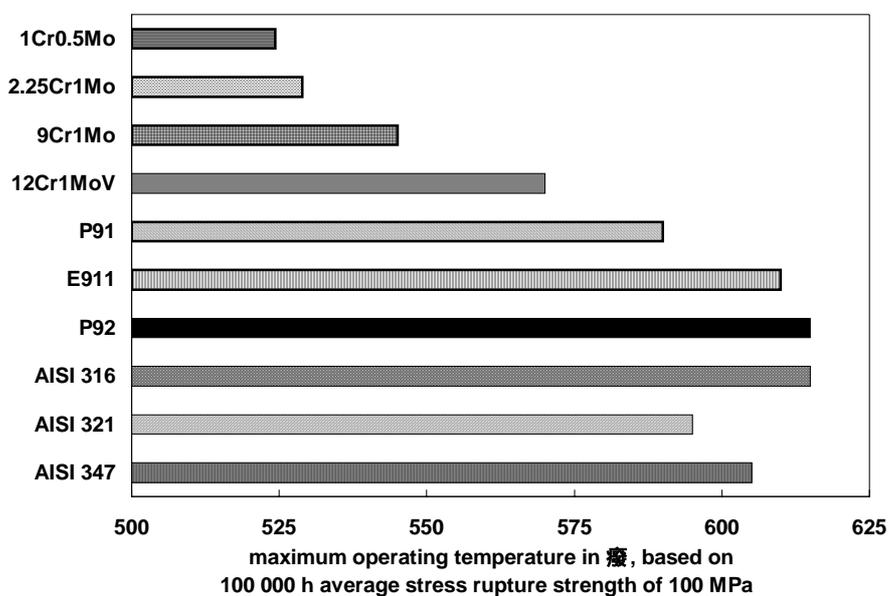


Figure 2 Maximum operating temperature for power station steels based on 100000 hours stress rupture strength of 100 MPa.

To determine the deformation behaviour of modified 9% Cr steels under upset conditions, investigations are underway at the Forschungszentrum Jülich (FZJ). Using the fire accident test specified for building materials, German Standard DIN 4102, the temperature at which catastrophic deformation occurs was determined for P92. The specimens were loaded and heated from room temperature at a rate of 600 °C/h and the strain measured. The stresses applied the specimens were 400, 300 and 100 MPa (80, 60 and 20 % of the room temperature yield stress, $R_{p0.2}$). The results are shown in Figure 5. At all three stresses, the material fractured before the ferrite/austenite transition temperature (A_{c1}) was reached. The maximum temperature that P92 would survive at 20% of its $R_{p0.2}$ is about 720°C and just under 500°C if loaded to 80% of its $R_{p0.2}$. When these temperatures at specified load are exceeded, the material deforms catastrophically.

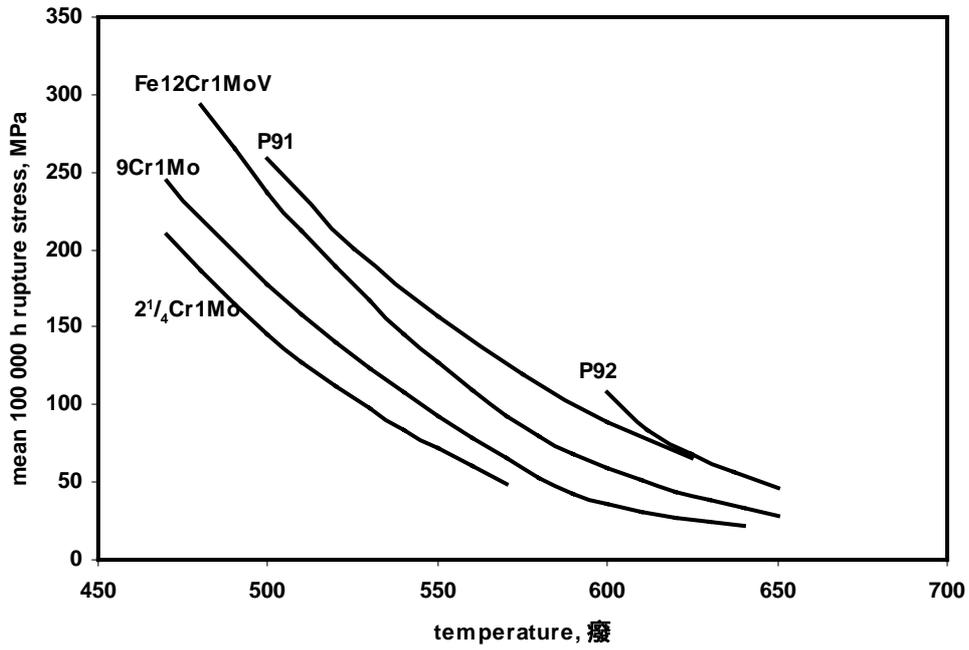


Figure 3 100 000 h stress rupture strengths of power station steels

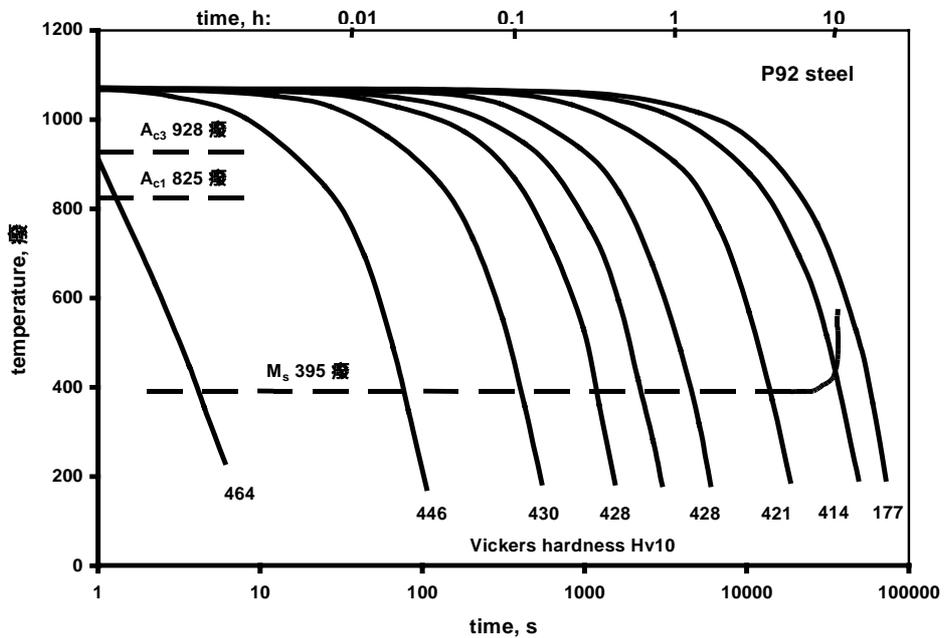


Figure 4 Time-temperature-transformation diagram for P92; the low hardness value of 177 indicates that, at the slowest cooling rate, the martensite transformation has not occurred

3. MATERIALS FOR THE HOT GAS DUCTS AND HEAT EXCHANGERS.

The principal candidates for these applications are the high temperature alloys, such as Alloy 617 (nickel-base) for temperatures above 850 °C and Alloy 800H (iron-base) for temperatures below 850 °C. The choice of the materials for hot ducting depends also on the thickness of the walls. Extensive characterisation of these alloys was carried out in the German HTR materials programmes, with emphasis on the stress rupture behaviour in simulated HTGR service atmospheres. The gas-metal interactions have been elucidated by Quadakkers and Schuster (1985) and indicated that carburisation or decarburisation could occur, depending among other factors on the materials and gas flow rates. Figure 6 shows a comparison of the stress rupture properties of Alloy 617 in air and in carburising HTR helium. It is noted from the figure that all the data, for large number of heats and product forms, form a scatter band for each temperature. The specimens tested in HTGR helium atmosphere exhibited high rates of carbon uptake, the carbon content reaching double the initial value after a few thousand of hours at 950°C. No appreciable effect creep strength, however appreciable effect on the tensile rupture strength was detected. However, carburisation significantly reduces the low temperature ductility, due to the presence of grain boundary carbide films (Lupton and Ennis, 1981). The low ductility temperature range was found to extend up to 800°C (Abdel-Azim et al 1990). From this aspect, carburisation effects must be avoided. It should be mentioned here that for all the mechanical testing done in the German HTR projects, the experimental data (in the case of creep rupture testing, the strain-rate data) has been preserved in the HTM Alloys Databank system developed at JRC Petten (Over), allowing further analyses of creep behaviour to be carried out.

With regard to the effect of decarburisation, the more likely interaction in service because of high helium flow rates, the creep strength was found to be dramatically reduced (von der Gracht et al 1986). Figure 7 compares creep curves for Alloy 617 in air, in carburising test gas and in decarburising gas. An additional test involved changing the test gas from carburising to decarburising during test. The reduction of creep strength is clear, illustrating the importance of grain boundary carbides for creep strength of Alloy 617. Decarburisation in HTR service must therefore be avoided.

The high cobalt content in Alloy 617 could pose a potential problem, although in German HTR projects, it was found that Co was not incorporated into the oxide scale and so radioactive Co would not therefore enter the hot gas circuit, even if the oxide spalled off.

4. MATERIALS FOR THE TURBINES

In the former German development programme for “Direct cycle HTGR”, the High Temperature Reactor with Helium Turbine (HHT) project, the main concerns for materials were related to:

- Rotor shaft
- Blades and vanes
- Hot ducts

The maximum helium temperature in the cycle was 850 °C. For all kinds of component the technological availability and fabricability was of high concern, then followed the material properties such as:

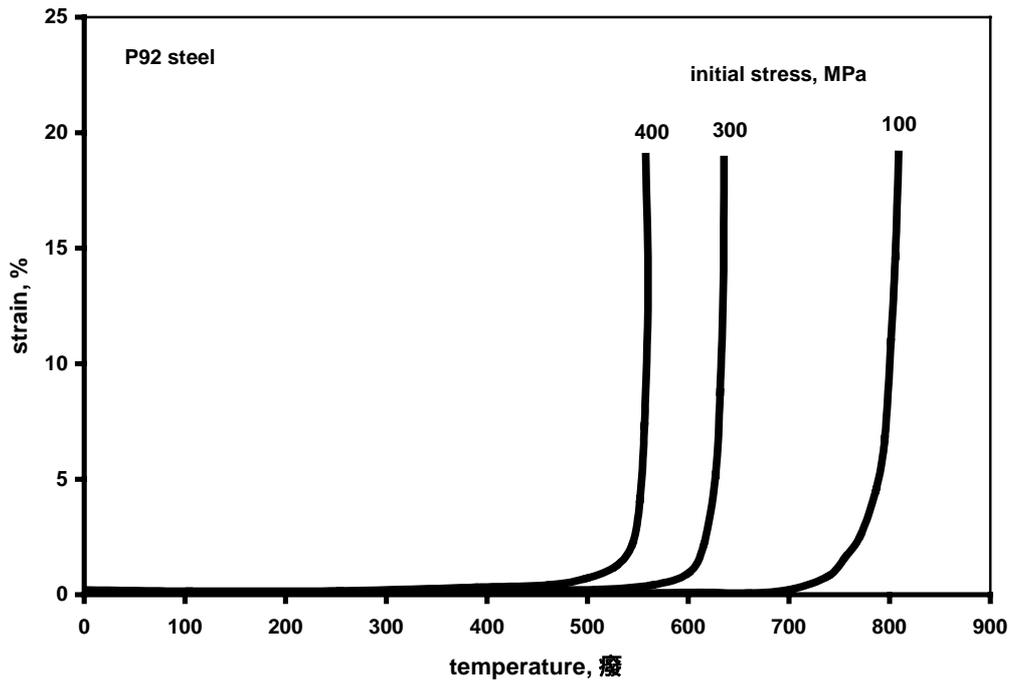


Figure 5 Fire accident test (DIN 4102) on P92, loaded and heated up at 600 C/h

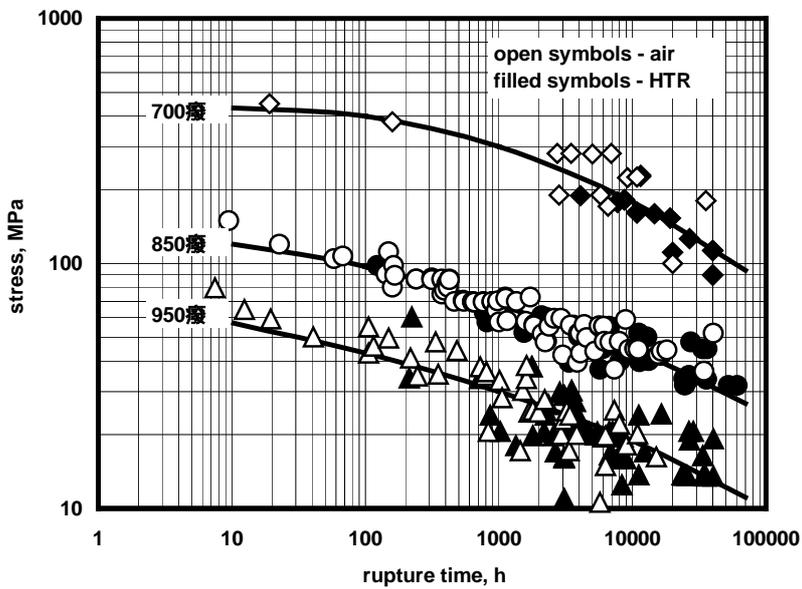


Figure 6 Stress rupture properties of Alloy 617 at 700, 850 and 950 °C tested in air and in simulated HTR helium atmosphere (carburising)

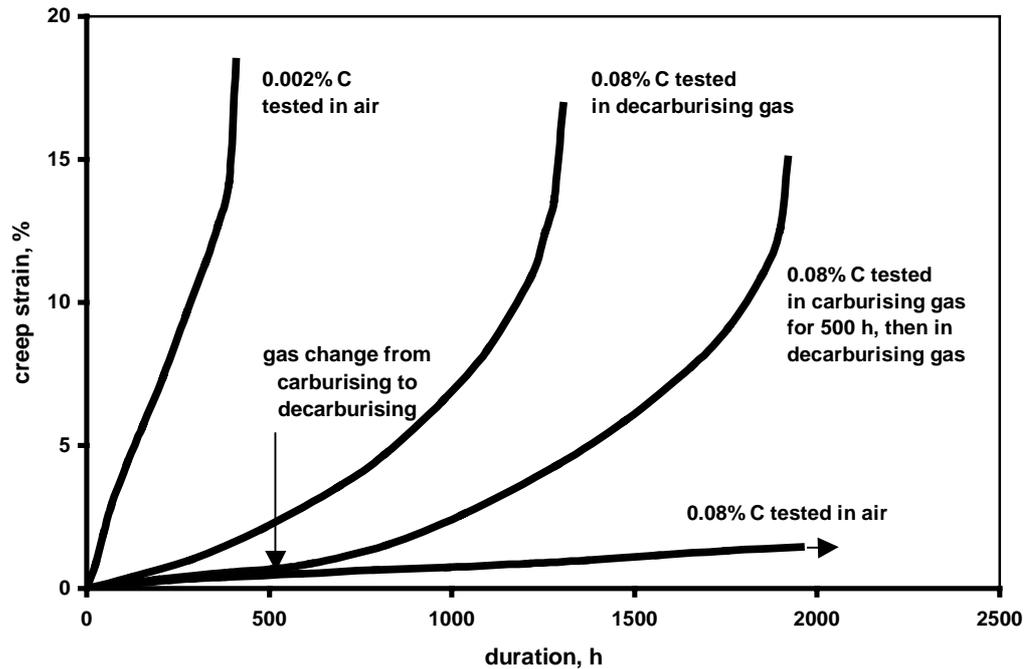


Figure 7 Creep curves for Alloy 617 tested at 900 °C, 18MPa: low C (0.002%) tested in air; normal C (0.08%) tested in decarburising gas (He-500 μ bar H_2 -1,5 μ bar H_2O); normal C tested initially in carburising gas (He-500 μ bar H_2 -100 μ bar CO -10 μ bar H_2O) and after 500 hours in decarburising gas; and normal C tested in air

- Temperature dependence of yield strength
- Temperature dependence of creep strength
- Fatigue resistance
- Effects of long term exposure under operational conditions (microstructural stability, RT-toughness, and for some alloys the ductile-brittle transition temperature, (DBTT)).

Especially for the rotor shafts, fracture mechanics, allowable structural inhomogenities, and NDT methods have been discussed. Each turbine must be maintained at given intervals, therefore for the nuclear coolant driven helium turbine questions concerning decontamination have to be solved. A survey of the achievements of this programme is presented in Nuclear Technology, volume 66, 1984. In industrial gas turbines cooling methods using air from the compressor can be used both for rotor shafts and discs as well as for blades and vanes. In closed loop helium turbines, such cooling will not possible.

4.1 Rotors

From a safety point of view, the design of rotor must be against burst (e.g. plastic collapse) due to overspeed of the rotor or disc. The safe and reliable service under operating conditions has to satisfy a number of design criteria, depending on the particular design. In any case, however, a rotor shaft or an assembly of rotor discs should never exceed a tangential speed at the rim which gives rise to stresses higher than the yield strength of the material at the highest operational temperature. The yield strength must also have a safety margin to avoid any kind of non-elastic deformation during all the operational duration. Over a complete start-up and shut-down cycle, the combined effect of the centrifugal and thermal loading induces a stress-strain cycle, which may locally be severe enough to initiate fatigue cracking or to allow cracks resulting from manufacturing defects (e.g. structural inhomogeneities) to propagate. For rotor discs, the maximum local stress range is found in the rim area at the root of the blade groove with a tentative stress concentration factor of $K_t \approx 2.5$ with the expected small strain range yields to an assumed lifetime of about 10000 operational cycles. However, if a manufacturing defect, which has remained undetected during non-destructive inspection, is located in a region of very high tensile stress, it may well grow to become critical. This is true, in particular, if the materials temperature in that location is high enough for creep crack propagation. Having in mind all of these requirements, the maximum temperatures of the once evaluated materials for rotors are:

- 21Cr Mo V 5 11(1% Cr Steel): 300 °C
- 12 Cr Ni MoV12 1 (12% Cr Steel): 335 °C
- 9 Cr Mo V Nb B (New COST Steel): 500 °C
- Inconel 706 (Modification of Inconel 718 for land based turbine): 600 °C

The production of large forgings (open die forgings) and the heat treatment, especially through hardenability, will be the main technological challenges for these components. Materials such as INCONEL 718, Waspaloy and Udimet 720LI, developed for aero jet gas turbines, have not yet been produced in the large diameters required for land-based gas turbines (Härkegård and Guédou 1998, Rösler et al 2002). To demonstrate the potential of these materials, we compare in Figure 8, the extrapolated 10^5 hours creep rupture strength of some of these materials.

4.2 Blades and Vanes

In the early eighties (HHT Project), following industrial gas turbine practice, cast Ni-base superalloys were considered for the blades and vanes in the first stages of the turbine, with wrought Ni-base alloys for the more moderate operating conditions in the last stages. In those days, an equiaxial conventionally cast materials such as Alloy 713LC were evaluated. Since in a helium turbine the cooling of blades and vanes will not be practicable, material temperatures should not be allowed to exceed 800 to 850 °C.

The primary stresses for the blades are about 150 MPa to 200 MPa, for all the duration at steady state operation. This requires a shorter operation time than that expected for the total plant lifespan. It is assumed that the blades of a helium turbine can be decontaminated to perform maintenance work at specified intervals, to enable, for example, replacement of the blades. The service life of the blades should be adjusted to the maintenance intervals. The candidate alloys for turbine blades included well known Ni-based alloys such as Alloy 713LC

(conventional) and NIMONIC 80A (closed die forged). In the later years of the German programme, newly developed precision cast (conventionally cast) Ni-base alloys were evaluated, in spite of the fact that these alloys contained Co as an important element which may lead to decontamination problems. The molybdenum-based alloy TZM, which cannot be used in open air gas turbines because of its catastrophic oxidation behaviour in air above ≈ 500 °C, was also considered for application in the helium turbine. The very high temperature strength of TZM can be only achieved in pure inert atmosphere, e.g. pure helium, with a very low oxygen partial pressure. Disadvantages of this alloy were low ductility at room temperature with a NDDT temperature above 300 °C and very high density.. Materials deterioration due to emergency air or water ingress was never solved for TZM (Jakobeit, 1984).

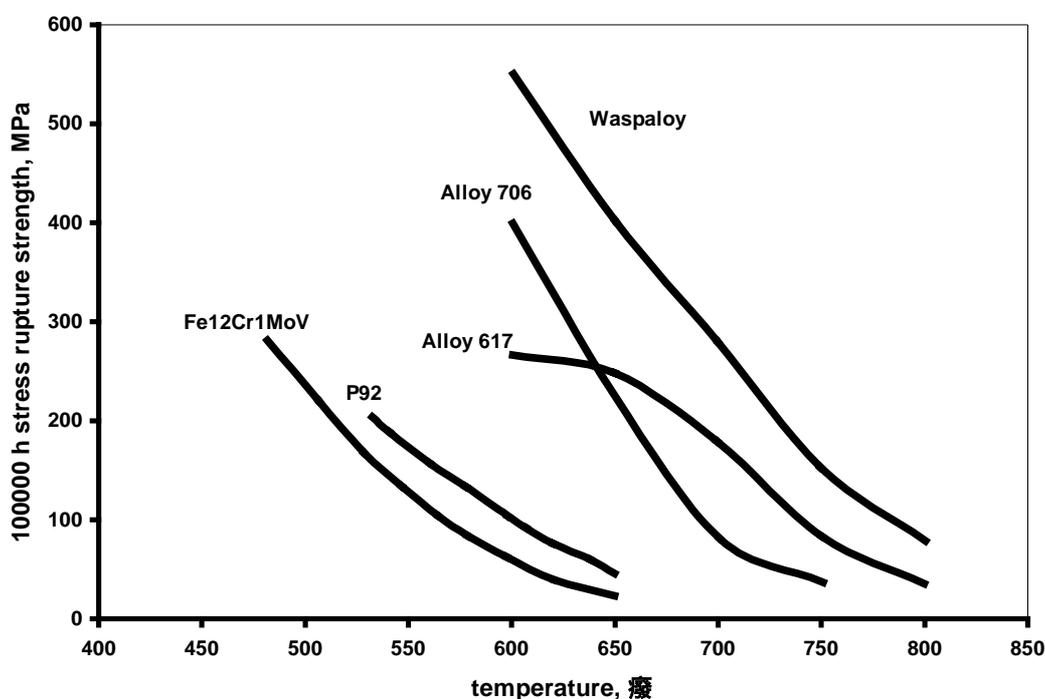


Figure 8 The 10⁵ h stress rupture strength of different alloys for rotor shaft, heat exchangers and hot ducting components (extrapolated)

In fossil fired steam and gas turbine power plants, higher operating temperatures have been achieved over the last few decades resulting in higher efficiencies. Besides the improvement of the cooling concepts for vanes and blades, the higher temperature metals have been achieved using alloy development for the introduction of directionally solidification and the single crystal technology. Additionally use of ceramic thermal barrier coatings allows combustion temperatures higher than 1350 °C but the metal surfaces temperatures are stilll restricted to 950 °C to 1000 °C, and, of course, cooling of the blades is necessary. In different COST programmes long term creep tests of new DS and single-crystal alloys up to about 50 000 hours have been determined; the very long term

experimental results are still missing. A first estimation of the potential of these materials in the conventional gas turbine technology can be derived from the temperature dependence of the yield strength given in Figure 9.

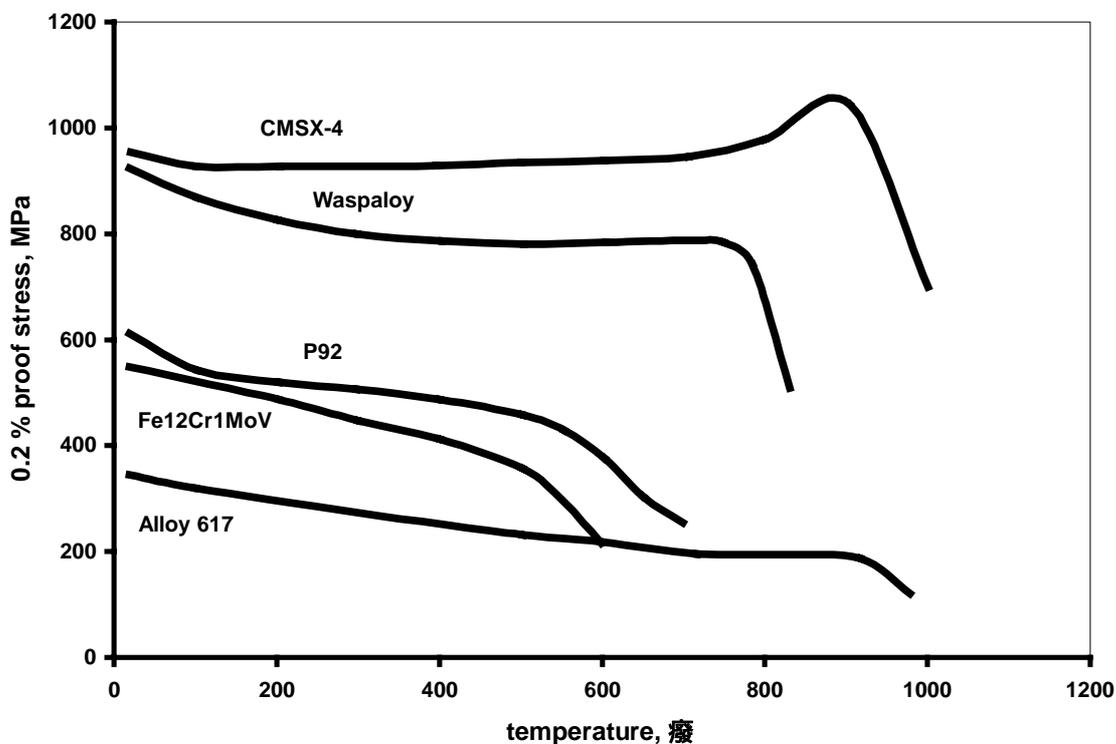


Figure 9 Proof stress of different materials as a function of test temperature

We note that the current European HTGR materials project (Buckthorpe et al 2003) has selected two turbine alloys for long-term evaluation for use in blades:

- IN 792 (Ni-0.05C-12.5 Cr-9 Co- 4.5 Ti -4W-4Ta-3Al-2Mo) conventionally cast
- CM 247 DS (Ni-0.15C -10 Co-4W-8 Cr-5.5- 3Ta-2Mo-1 Ti) directionally solidified

It must be borne in mind that for both alloys there remains the question of Co and the release of radioactive Co-containing particles in the primary circuit that may lead to problems in maintenance.

Figure 10 summarises the 1000 h stress rupture strengths of turbine blade materials currently under investigation, compared with the alloy earlier selected in the German HHT programme, IN 713 LC. The lack of long-term stress rupture data for the newer alloys means that a reliable estimation of the stress rupture strength for service relevant durations cannot yet be made.

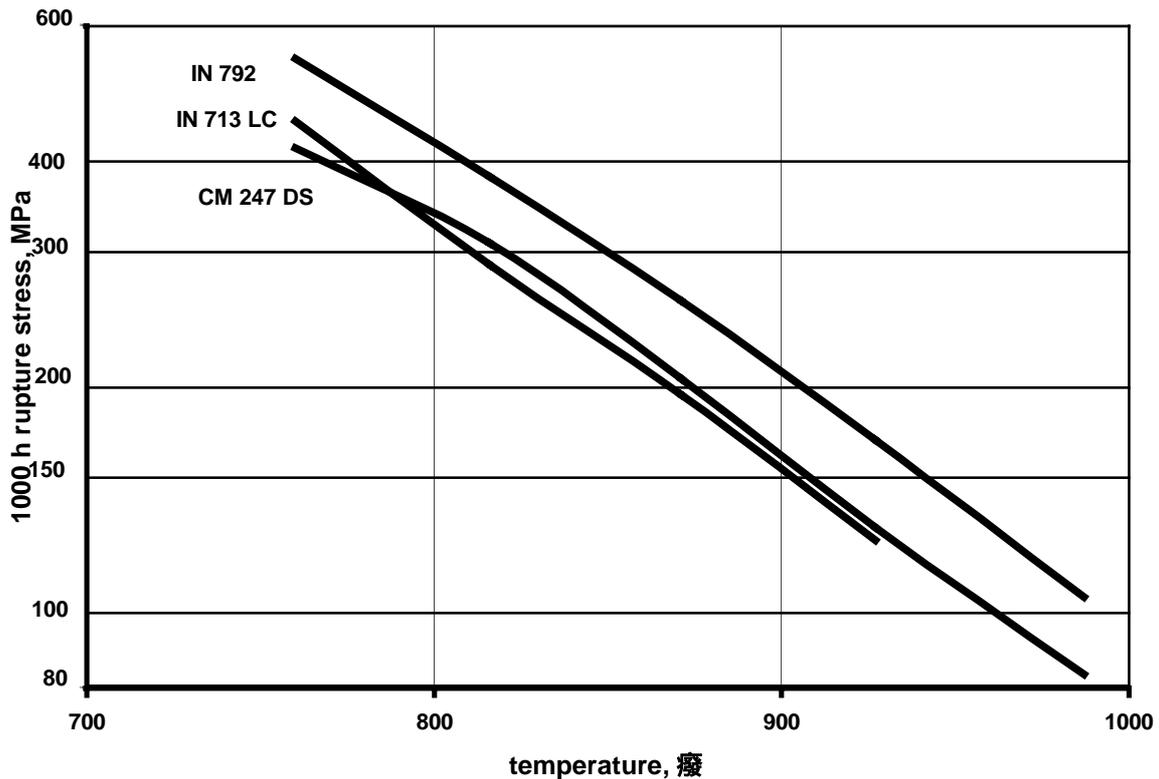


Figure 10 1000 hour stress rupture strength of different alloys for turbine blades

5. CONCLUSIONS

The selection of material for the structural components of an advanced HTGR is determined by:

- the expected highest materials temperatures
- the long-term creep resistance at those temperatures
- long-term structural stability.

In all cases, the structural materials should not be exposed to high flux of either fast or thermal neutrons. The higher the temperature, the more sensitive the structural materials become for loss of deformability due to neutron irradiation (thermal neutrons). Based on the considerable experience from HTGR materials programmes carried out in Japan, US and Germany in the eighties, structural materials have been evaluated and can meet the requirements for use in HTGR with the helium gas turbine or in the heat exchanger for helium temperature not higher than 950°C. Higher temperatures will require an enormous amount of additional research for materials. For heat exchanger materials, gas-metal interactions are of great importance. Both decarburization and carburisation can occur in impure helium, but can be kept within acceptable limits by careful control of the impurity levels in the helium.

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