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MATERIAL DEVELOPMENTS FOR SUPERCRITICAL BOILERS AND PIPEWORK – BRIDGING THE GAP ?

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Abstract

This paper briefly reviews the current state of developments for ferritic, austenitic and nickel alloys for supercritical steam raising power plant applications. Some of the challenges are touched upon as well as various activities ongoing to meet these challenges. It is likely that, although the technical challenges are significant, developments in ferritic alloys will allow a more incremental increase in plant efficiency than the step change required to justify the cost of nickel alloys. This conclusion may change as the cost of fuel increases such that the economic and commercial risks of using nickel are compensated by the saving in fuel costs.

Key Words: Boilers, Ferritics, Austenitics, Nickel Alloys.

1 INTRODUCTION

The drive for ever-increasing efficiency of power plant has always been present but in recent years it has been given greater impetus by the concerns over global warming, particularly in relation to the emission of the greenhouse gas carbon dioxide. Apart from simple economics demanding lower electrical energy unit costs, there is the need to reduce the amount of emissions per megawatt generated, summarised in figure 1. This also suggests that the main contributor to emissions reduction will be plant efficiency [1]. The maximum available thermal efficiency of power plant is that of a reversible device absorbing thermal energy by heat transfer at the source temperature and rejecting thermal energy at the sink temperature, in which case

$$\text{Efficiency} = 1 - (T\ \text{sink}/T\ \text{source})$$

The sink temperature is normally fixed depending on geographical location leaving the source temperature as the major variable in the quest for higher efficiency. No real cycle is truly reversible, and when this is taken into consideration efficiency is also a function of pressure. In particular, when the pressure is above the critical pressure i.e. supercritical, efficiency improvement is gained by avoiding the latent heat of evaporation.

For example, an increase in conditions from 305bar at 585/602°C to 320bar at 620/640°C will result in an overall increase in efficiency estimated at about 6% [2]. The gradual increase in temperature and pressure is shown diagrammatically in figure 2, which also indicates the target for 2020 [3]. The step change in the target for 2020 results from the move from thick section ferritic headers and pipework to nickel alloys and their higher capabilities as determined through the AD700 European initiative. Because of the resultant significant increase in material costs, it would be necessary to maximise the efficiency by operating at as high a temperature and pressure as practicable.

This step change is more radical than the usual incremental improvements traditionally seen in the boiler industry and would inevitably be technically challenging. Nevertheless a project known as EON50+ was started in 2008 with the objective of achieving in excess of 50% net plant efficiency (%LHV basis) at final steam conditions around 700°C and 350 bar. Several boiler makers developed outline designs with a view to plant operation around 2015 but due to the economic downturn the project is currently shelved. However, the need for efficiency improvements is driven, in part, by the energy penalty for CCS (Carbon Capture and Storage), see figure 3, with estimates ranging from 5 to 8% efficiency loss for the implementation of CCS [1].

Increasing the temperature and pressure increases the demand for higher creep strength and steam oxidation resistant materials. In addition to these attributes the most suitable material types can vary depending on the application and fabrication or performance requirements. These can be divided into three areas –

- Waterwalls should preferably be fabricated without the requirement for post weld heat treatment.
- Other heat transfer tubing experiences a range of temperatures up to the highest metal temperatures in the final superheater section.
- Headers and manifolds are the thickest components and require good fatigue and creep/fatigue performance to accommodate thermal stresses during heating/cooling. This is related to the thermal conductivity and coefficient of expansion of the material.

On this basis for the thick section piping components the current best in class Creep Strength Enhanced Ferritic (CSEF) steel, P92, has an upper limit in non-heat transfer applications of ~610°C primarily due to the steam oxidation consideration. For the same reason it is limited to even lower temperatures when heat flux is relevant as in superheater elements or waterwalls. For thin section applications it is possible to move to the higher chromium alloys such as austenitic stainless steels but in thick section, due to poorer thermal conductivity and high thermal expansion of this class of material this is currently not considered practicable. The simplest technical solution for temperatures in excess of ~610°C is the use of nickel based superalloys, see figure 4, which have physical properties closer to the ferritic/martensitic alloys than the austenitic alloys. Unfortunately the cost of the nickel materials is very high and the increased demand in the market would place a strain on the supply chain. Furthermore the effects of fabrication and welding on long term properties require more comprehensive evaluation and there is limited field experience. Consequently the commercial cost and risks involved makes the use of nickel based alloys somewhat less attractive.

One of the fundamental questions that need to be addressed is: can higher efficiency plant be achieved without the need or the limited use of nickel based alloys. Figure 3 suggests that efficiency losses can be reduced by improvements in the CCS process. Similarly plant improvements can assist in contributing to overall plant efficiency [4]. The real questions for the materials community are can ferritic materials be used above 610°C, or can the application range of austenitic stainless steel alloys be extended and thereby limit the need for the expensive nickel alloys?

In essence, can the gap between the best available CSEF alloy grade 92 and the very expensive nickel based alloys be bridged and thus enable a more incremental and hence lower risk approach to improving power plant efficiency? In this review some of the issues are considered for each class of metal that will affect the answer to this question.

2 FERRITIC/MARTENSITIC ALLOYS

Fleming et al [5] and Viswanathan & Bakker [6] provide a good review of developments for ferritic alloys up to the year 2000 for thick section components. At the time P91 was the industry standard for thick section components. Since then the nominally 2% tungsten alloyed variant P92 has become more popular due to its higher creep strength. The 1% tungsten alloy (E911) developed in a European Programme (COST 522) has not proved so popular with its creep strength intermediate between grade 91 and grade 92. All of these alloys suffer from in service issues relating to type IV cracking, and other critical factors such as low N:Al ratios and inferior martensitic structure due to criticality of the thermal processing parameters [7][8]. Both grade 92 and E911 with W additions have been approved in European and ASME codes, but their allowable stresses have been progressively reduced due to the continuing accumulation of long term data. It has been considered that this has been contributed to by the weakening effect of Laves phase $(Fe,Cr)_2(Mo,W)$ formation [9] which reduced the solid solution strengthening contribution of tungsten. Another potential issue with the 9%Cr alloys is their lack of steam oxidation resistance as the operating temperature exceeds 610ºC in pipework applications, as indicated in figure 5 [10].

For many years higher Cr alloys have been evaluated without success primarily due to the formation of a coarse Z-Phase, where the chromium combines with the strengthening MX precipitates containing niobium, vanadium and nitrogen, [11] thereby removing them from the microstructure. This generally occurs at extended time and hence the effect is not noted till a considerable proportion of the creep life is exhausted after which a sudden drop in properties is experienced. Current developments in creep enhancing this class of alloy are centred on the MarBN philosophy developed by F.Abe or using controlled precipitation of Z-phase as the strengthening mechanism as proposed by J.Hald.

MarBN uses controlled additions of Boron and Nitrogen to the Martensitic matrix but care must be taken with the latter to avoid formation of boron nitride which is detrimental to the creep properties [12]. This can result in an improvement in creep life for a 9% Cr alloy of up to 50% in stress compared with grade 92. It has also been reported that the boron prevents grain refinement within the heat affected zone and should thus reduce or eliminate the type IV problem prevalent in this type of alloy [13]. However obtaining a matching weld metal, with the required close control of the chemistry especially nitrogen and boron, may prove problematic with one possible option being to use overmatched nickel filler materials. It is also perhaps not surprising that weld metals, essentially a series of small castings and only subjected to post weld tempering, fail to demonstrate matching creep strength for these types of alloys, bearing in mind the critical thermal cycles applied to the CSEF base metals to fully develop their long term properties.

Danielsen and Hald have studied the deleterious effect of Z-phase [14] in some detail and support the mechanism of the precipitation of non strengthening coarse Z-phase at the expense of strengthening MX particles and consider this to be the main reason for the inability to develop higher strength 11 – 12% Cr steels. An experimental steel has been produced based on the concept of accelerating the Z-phase formation by chemical composition control in order to suppress deleterious growth [15], shown schematically in figure 6. This 12Cr-0.2N-0.2(V+Nb)-balance Fe material has been demonstrated to cause each MN particle to turn in to one Z phase particle, and be immune to the dissolution of strengthening nitrides. Creep tests on the alloy are currently underway.

These two main avenues of research are focussed upon creep strength enhancement potentially resulting in adequate creep strength (100MPa – 100khrs) up to 650°C, but there is still the question regarding steam oxidation resistance particularly for the 9% Cr MarBN type. The main routes to enhance steam oxidation other than chromium additions have been to enhance the chromium diffusion and surface coating. For austenitic superheater and reheater tubes fine grain structures and cold working the surfaces have been used to enhance chromium diffusion and hence the rapid development of the protective magnetite layer. This is discussed in more detail in the “Austenitic Alloys” section. The current ferritic materials e.g. grade 91 are already considered to be fine grained, typically ASTM 6 or finer. The effect of cold work, e.g. peening and the resultant grain refinement, has been shown to enhance chromium diffusion [16] and could be further explored.

Coatings have the potential to give long term benefits in terms of protection from steam oxidation, [17] [18] see figure 7. Many coating compositions have been trialled using chemical vapour deposition (CVD), slurry and pack cementation techniques. The primary aim of the coating process is to form a sufficiently thick Fe_2Al_5 so that the stable oxide formed is Al_2O_3 and a sufficient reservoir of aluminium for the expected service duration, see figure 8 [19] [20]. The long term microstructural stability and mechanical integrity of slurry deposited coatings on P91 and P92 have been evaluated in SUPERCOAT; an EU funded 5th Frame Work Project. The main conclusions from this work are “*Slurry aluminide diffusion coatings are protective for at least 41,000 h but degrade by coating-substrate interdiffusion leading to a decrease on the Al concentration at the surface, at the deposition of AlN precipitates within the substrate and to the development of Kirkendall porosity at the coating-substrate interface. However, based on long term steam oxidation experimental data, the life of the coating can be estimated to surpass 78,000 h.*” [17]

3 AUSTENITIC ALLOYS

Figures 4 and 5 summarise the advantage the austenitic alloys have over the ferritic alloys, namely improved creep strength and with typically 18%Cr a greater resistance to steam oxidation. The super austenitic alloys have even greater oxidation resistance with HR3C containing 25%Cr and Sanicro 25 containing with 22.5%Cr. Even though there is a cost penalty for using the austenitic alloys relative to the ferritic alloys the main issue is that of $\alpha\Delta T$, i.e. thermal fatigue. This family of alloys have a low thermal conductivity, typically 16W/m°C vs. 50W/m°C, and high thermal expansion, typically $18x10^{-6}$ vs. $11x10^{-6}$, relative to the ferritic alloys. For these reasons austenitic alloys are currently restricted to the superheater and reheater tubing and seldom for thick section components, a notable exception being the stub headers at Drax Power Station, UK which employed 15%Cr10%Ni6%Mn alloy (1.4982) the most creep strong austenitic alloy at that time circa 1975.

A recent review of service experience has been conducted by Shingledecker and can be found in reference 21. Of particular note in this review were the tests conducted at Eddystone Unit 1 where 6 different tubes were run for 13 years (75,075 hrs) nominally at 615°C and 35 MPa where the higher chromium containing alloys showing little sign of either steam or fireside corrosion, with the fine grained 18%Cr 347HFG performing as well as the 21%Cr Tempalloy A3. Post service samples of 304H from Callide C3 and C4 units tested in stress rupture showed minimal reduction in life expectancy relative to virgin material even after 80,000 hours service, see figure 9.

The improved creep strength of the current super austenitic alloys would mean that, for the same steam conditions thinner sections would be possible compared with the conventional austenitic alloys e.g. 304H and 316H making pipe sized components feasible. In particular the recently developed Sandvik alloy Sanicro 25, which exhibits 100khrs average rupture life at 700°C and almost 100MPa, would seem like a good candidate. It has also been reported that ORNL together with Caterpillar are currently assessing the creep-fatigue performance of a cast alloy CF8C-plus in high temperature thick section applications [22]. If such materials could be used safely this would help to bridge the gap between the current best available ferritic alloys and the very expensive nickel alloys and permit incremental improvements in steam conditions and efficiencies.

The development of the advanced austenitic alloys has come about partly by specialised thermo-mechanical treatment but mainly by the addition and optimisation of alloying elements in particular niobium [23]. This forms fine NbX precipitates both with carbon and nitrogen and also contributes to Z-phase all of which greatly increase the creep strength [21]. However there is evidence that this also has the greatest influence on Strain Induced Precipitation Hardening. This results from cold working the material for example during bending and subsequent thermal exposure in the temperature range 540°C to 750°C. This can result in reduced creep ductility and premature failure. This carbide precipitation can also lead to reduced impact toughness [21] which becomes more significant for the higher chromium containing alloys [24]. For the HR3C type alloy Peng et al.[24] show that at 700°C both grain boundary and intra grain $M_{23}C_6$ precipitate out within 1000 hrs with a huge reduction in toughness, see figure 10.

Steam oxidation of 18/8 type austenitic tubes has proven to be of concern in many recently commissioned boilers with excessive growth of the bore oxide leading to tube blockage and failure and /or turbine erosion.

Steam oxidation problems can be reduced by four main methods:

- Employing a higher Cr content alloy.
- Chemical alteration of the surface by, for example chromising.
- Employing fine grained materials.
- Shot peening of the surface.

The first two options are obviously expensive, and from that point of view the latter two options would appear more attractive. The logic behind the fine grained structure of an alloy such TP347HFG is based on increasing the diffusion of chromium in the alloy by considerably increasing the number of grain boundaries. Grain boundaries are regions of high disorder and vacancies which allow the substitutional chromium atoms low energy sites and also “corridors” to the surface where they can lower the energy of the system even more by forming oxide. So TP347HFG has enhanced oxidation resistance in comparison to TP 347H by virtue of having a higher chromium level in its scale. Cold working, by introducing slip lines and point defects, also promotes Cr diffusion [25] giving rise to the same protective mechanism as fine grained material. Both methods of enhancing Cr transportation make a considerable difference to scale growth as can be noted in figure 11.

Welding of the austenitic stainless alloys is fairly straightforward with the same welding processes being generally available as for the ferritic steels. However these alloys are austenitic at room temperature and do not go through the phase transformation on cooling which the ferritic/martensitic alloys do hence cooling rate from welding is not critical. Consequently preheating is neither required nor desired but on the other hand heat input and interpass temperature should normally be more controlled to avoid solidification cracking issues particularly for the fully austenitic grades [26]. As mentioned previously the alloys have a low thermal conductivity and high coefficient of thermal expansion which combine to make them more susceptible to distortion. Inspectability by the ultrasonic technique, which is often carried out both pre-service and in-service, can prove difficult due to the epitaxial grain structure causing high attenuation and beam deflection. For this reason on thick section components many fabricators favour narrow gap reduced weld volume welding techniques. This also brings obvious economic benefits after the purchase of the equipment and helps to de-skill the operation resulting in more consistent, defect free welds with minimum distortion.

It may be concluded that up to circa 650°C for tube applications austenitic alloys could perform satisfactorily, but above this temperature or for thick sections much work needs to be performed. One active area of research is the alumina forming austenitic alloys where at temperatures above 600°C the alumina scale is more adherent and protective than the chromia scale formed with low aluminium containing austenitic alloys [27] [28]. These alloys are strengthened by MC, primarily NbC, and NiAl intermetallics that are very stable at elevated temperatures. Laves phase, $Fe_2(Mo,Nb)$, formation is controlled by careful control of the Mo and Nb content [28]. The use of the alumina forming austenitic alloys may allow their use up to 700°C, although for thick section there still remains the thermal expansion issue. This was touched on by Perrin and Fishburn where they recommended the need to review the ASME Boiler and Pressure Vessel Code and the Water Tube Boiler Code, EN12952 to account for thermal cycles [29].

4 NICKEL ALLOYS

High creep strength nickel alloys were originally developed for the aeroengine industry leading to significant improvements in performance [30]. These alloys are often highly alloyed with exacting thermal cycles required to develop the optimum properties for the relatively short lived components. For the power generation industry a few of these aeroengine alloys are being evaluated along with a new generation of alloys that have been developed where fabrication and long term creep are the primary considerations [31]. The current use of nickel alloys in pressure parts of power plant is limited but the advantages in terms of creep, see fig 4, and oxidation resistance [32] over the austenitic alloys are driving the interest in these alloys. There are factors other than creep and oxidation resistance, however, which have to be taken into consideration.

The nickel alloys being FCC materials can suffer the same propensity to ductility dip cracking (DDC) as the ferrous based austenitic alloys [33] [34]. DDC is a grain boundary sliding creep-like phenomenon principally affecting welds and heat affected zones. DDC is linked to grain size, impurity segregation, grain boundary tortuosity, and intergranular precipitation. If welded under high constraint conditions the available ductility may be exceeded by the welding induced strain, i.e. contraction, leading to DDC. With a higher coefficient of expansion relative to iron, nickel alloys need careful handling during welding. Ramirez et al [33] advocate the controlled additions of Nb and Ti, strong carbide and nitride formers, to the welding consumable to promote grain boundary tortuosity via grain boundary precipitation.

A similar mechanism variously called relaxation cracking; stress induced cracking; strain age cracking etc., again a grain boundary creep-like phenomenon, involves the re-precipitation of phases dissolved during welding [35]. At critical temperatures very fine intergranular carbides rapidly precipitate acting as dislocation pins in the intergranular regions, whilst grain boundary carbide growth absorbs any local fine carbides leading to channels of weak material along the grain boundaries. Standard tests have been developed to assess the susceptibility of nickel alloys to relaxation cracking e.g. the controlled heating rate tensile test (CHRT) [36] see figure 12. This shows that the high γ' forming alloys, i.e. high Al and Ti, are more prone to relaxation cracking than the low γ' forming alloys making these attractive for power plant applications.

Woodford's review on Gas Phase Embrittlement in aeroengine nickel alloys [37] suggests that consideration to environmental grain boundary attack needs to be considered for power plant nickel alloys. It is suggested that time dependent intergranular crack propagation in air under either sustained or cyclic stress is a result principally of oxygen attack ahead of the crack tip and that any separate effect of time dependent deformation (creep) is very small relative to the environmental effect. The ability to form a stable oxide scale can reduce or eliminate oxygen damage and time dependent cracking. The interaction of environment and applied loading in service is a further complication, see figure 13. Although the test conditions are severe for power plant applications, the data provide indications for plant operation. Figure 13 shows that life expectancy is reduced when oxygen is present, but of more interest is the observation that the life expectancy for a dwell of 18 and 1800 seconds are the same, i.e. there may be no creep fatigue interaction [38]. This observation is also confirmed by Hu and Wang with their work on super Waspaloy at 600°C [39].

For applications where high creep strength is required the γ' forming materials are necessary where control and stability of this phase is critical i.e. the greater the volume fraction and number of particles the greater the creep strength [40]. Coarsening of the γ' phase, and hence reduction in creep strength, is controlled by diffusion of Al and Ti through the Ni matrix [41] and very dependent upon temperature, see figure 14. Consideration of the coarsening rate and dissolution temperature of the γ' phase is a major consideration in deciding the upper operating temperature of an alloy. For thick section components, the reduced cooling rate from the solution treatment temperature can prevent ideal conditions for γ' precipitation. Mitchel et al [42] working on aeroengine alloys found that reducing the cooling rate produced a bimodal γ' distribution with increased precipitate size. It was also noted that slow cooling rates generate low γ-γ' mismatch which in turn may promote γ' instability during subsequent ageing heat treatments or during prolonged in-service exposure.

5 CONCLUSIONS

Some of the factors driving the need for higher efficiency in power plant cycles have been reviewed and the role of materials developments and the nature of these developments in relation to steam raising plant components have been considered.

In summary:

- There is a simple economic driver to increase cycle efficiency and this will become greater with the incorporation of carbon capture technologies into plant designs.
- The highest thermal efficiency in presently operating plant is achieved with final steam temperatures around 610°C and this limit is dictated by the strongest available CSEF alloy P92 employed in thick section components such as headers, manifolds and main steam pipework.
- Designs of plant with final steam temperatures of 700°C have been developed (AD700 programme). Construction of such plant would result in an historically unprecedented increment in cycle efficiency only made possible by the use of nickel alloys.
- Nickel alloys bring with them a different catalogue of potential integrity threatening mechanisms without any significant plant experience and introduce a level of risk. For these reasons, smaller increments in efficiency brought about by moderate improvements in ferrous alloys, may be preferred.

It is considered that the following areas of development can help bridge the gap between the current steel based alloys and the nickel superalloys.

- Mechanisms to increase the creep strength of the 9-12%Cr steels are currently under active investigation; these include boron containing steels and steels incorporating chemical composition restrictions to alter the precipitation mechanism of the normally deleterious Z-phase.
- Enhanced creep strength of 9-12%Cr steels could elevate the permissible operating temperature to a level in excess of their steam oxidation or corrosion capabilities creating the need to develop suitable internal and external coating technologies.
- Creep stronger and more oxidation resistant austenitic alloys have been developed for thin section components where the intrinsic low thermal conductivity and high thermal expansion are of little consequence. It may be possible, by design, to extend the application of these to thicker components.
- Thick section austenitic stainless steels however, may require controlled start up and shut down cycles restricting them to base load duties. With the advent of significant renewable energy production it is likely that the need for a more flexible grid will become paramount.

Hence these developments may allow a more incremental increase in plant efficiency than the step change required to justify the high cost of nickel alloys. This conclusion may change as the cost of fuel increases such that the economic and commercial risks of using nickel are compensated by the saving in fuel costs.

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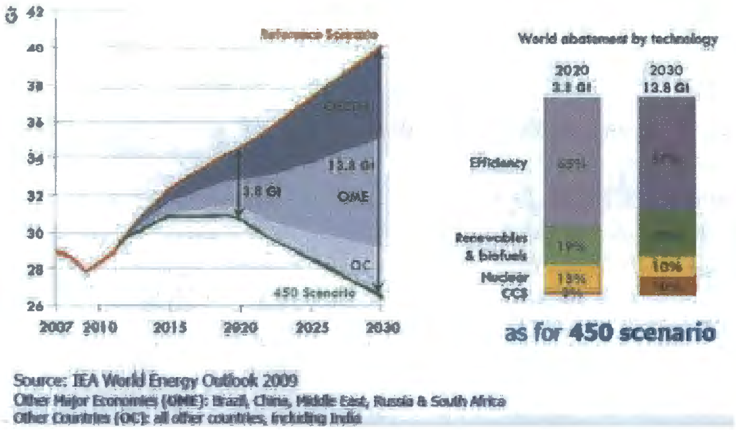


Source: IEA World Energy Outlook 2009
Other Major Economies (OME): Brazil, China, Middle East, Russia & South Africa
Other Countries (OC): all other countries, including India

Fig. 1:- The Driver for Higher efficiency Power Plant (1)

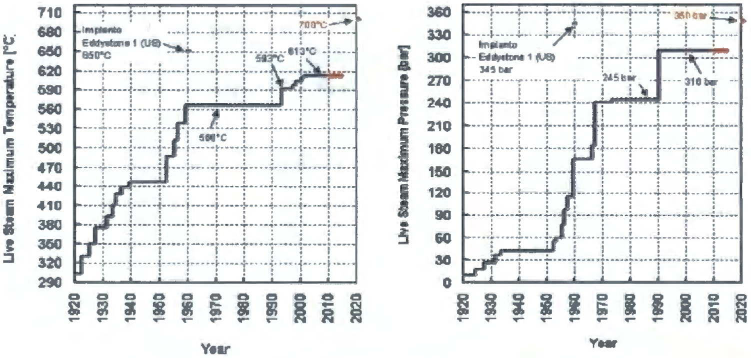


Fig. 2:- Change in steam temperature and pressure required for HSC plant (3)

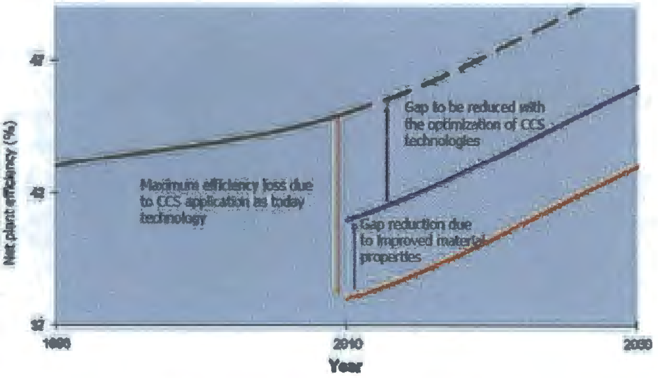
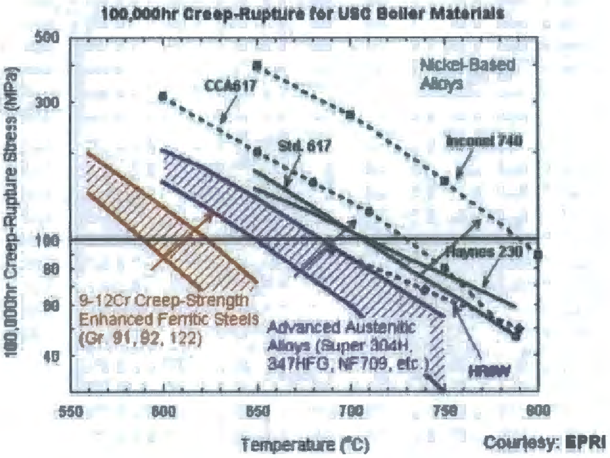


Fig. 3:- The Energy Penalty for CCS (1)



Courtesy: EPRI

Fig. 4:- Comparison of creep strength for different classes of metals.

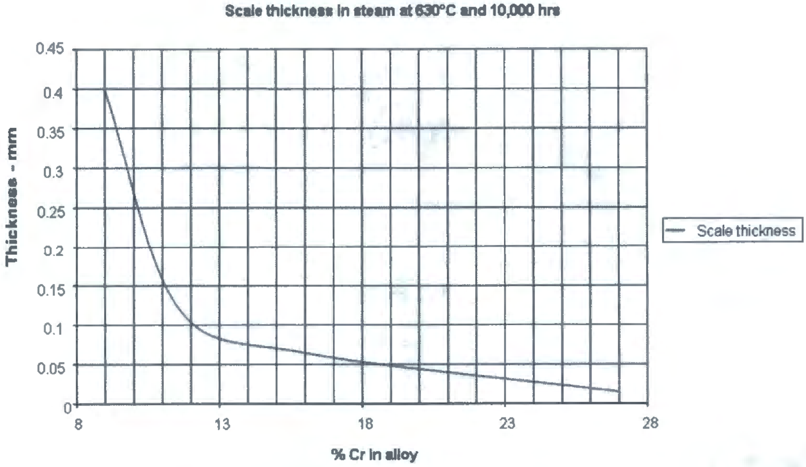


Fig. 5:- Effect of Cr content on steam oxidation resistance (10).

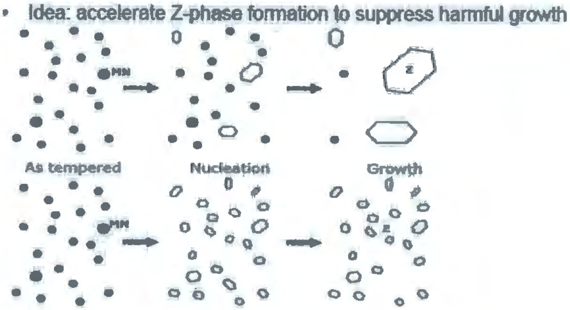


Fig. 6:- Schematic of Z-phase strengthening concept, top sequence shows MN strengthened material and bottom sequence Z strengthened material (11).

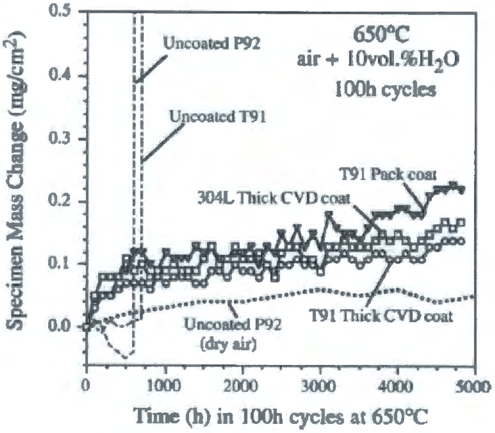


Fig. 7:- Potential benefits of aluminide coatings (17).

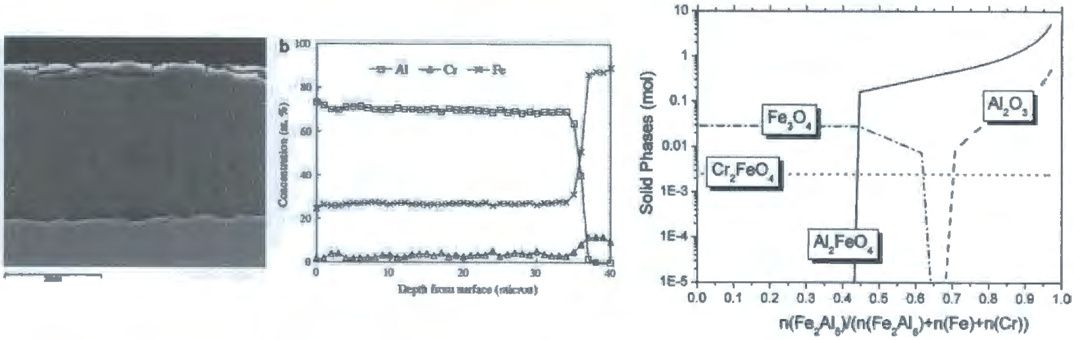


Fig. 8:- Micrograph of pack coating deposited on P92, left, with composition profile, middle (19). Thermodynamic calculation, showing the stable oxide phases formed under steam oxidation conditions (18).

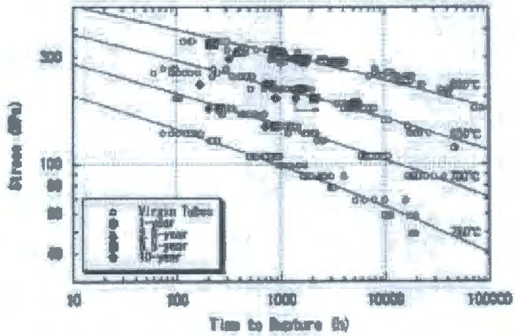


Fig. 9:- Stress-rupture properties of Super 304H tubes removed at various times from an industrial boiler (closed symbols) compared to virgin material (open symbols) for times up to ~80,000 hours (10 years) (21)

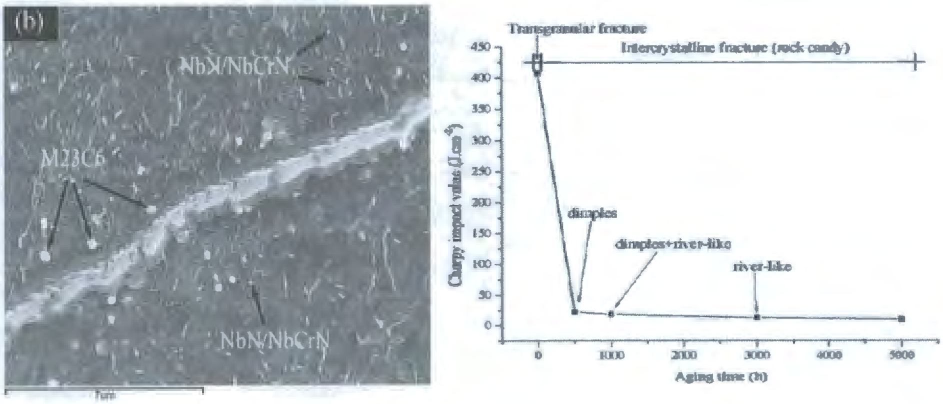


Fig. 10:- HR3C type alloy aged at 700°C. Precipitation of carbides and nitrides after 500hrs, left, and associated reduction in impact energy, right (24)

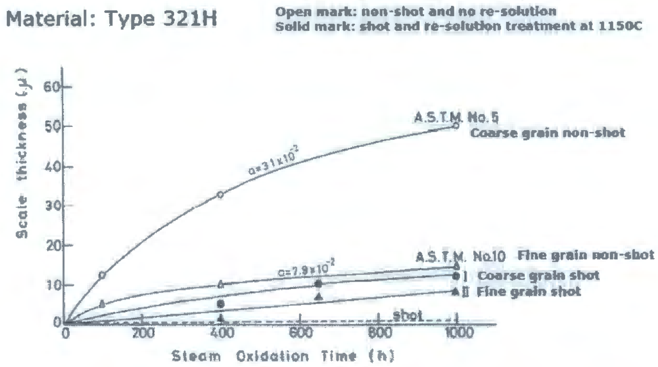


Fig. 11:- Effect of grain size and shot blasting on steam oxidation of 321H at 600°C (25)

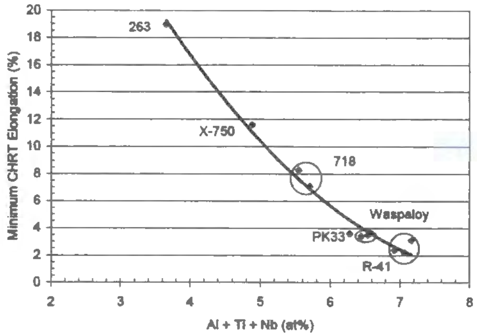


Fig. 12:- CHRT minimum elongation as a function of the total atomic percentage of precipitation-strengthening elements in a set of commercial alloys (36).

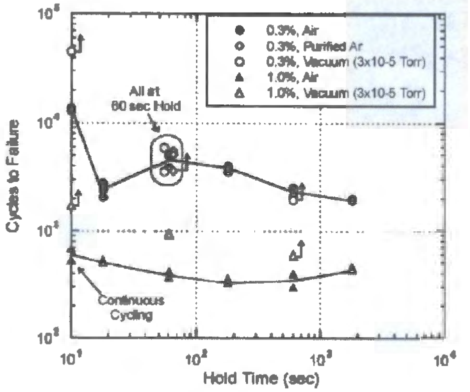


Fig. 13:- Alloy 617 creep-fatigue life at 1000°C as a function of hold time (38).

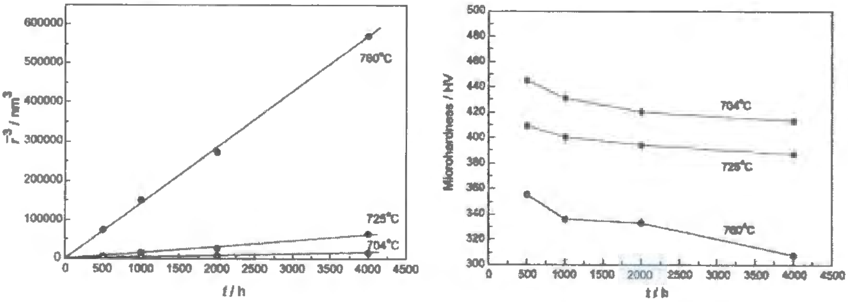


Fig. 14:- Coarsening of γ' precipitates in a 740 type alloy, left, and the corresponding effect on hardness, right (41).