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Use of Surface Modification of Alloys for Ultrasupercritical Coal-fired Boilers

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ABSTRACT

Ultrasupercritical (USC) coal-fired boilers, currently under study, will be required to utilize a variety of new, high strength alloys. These alloys will have improved mechanical properties compared with more traditional boiler materials and so will be suitable for operation in higher temperature service. However, environmental resistance, i.e. internal steam oxidation and external coal-ash corrosion, will be a factor limiting application of some materials under consideration. In those cases, the operating range of lower-cost alloys can be significantly extended by the use of surface modification techniques. This paper will review potential surface modification techniques and report on early test results of some laboratory evaluations.

Introduction

The implementation of ultrasupercritical boilers requires materials with high-temperature creep properties and high-temperature oxidation and corrosion resistance. New ferritic, austenitic and nickel-base alloys have been designed to meet the creep resistance demands, but the high operating temperature poses the risk of accelerated material degradation in various harsh environments. Both fireside corrosion and accelerated oxidation at elevated steam temperatures are of concern.

Surface modification techniques provide an alternative to the very costly more highly alloyed materials. The science of thermal spray has evolved in the last 15 years with the implementation

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of high-density spray techniques, such as High Velocity OxyFuel (HVOF) that have improved the quality of the applied coatings. Cold spray is another emerging technology that, combined with nano-size powders, can provide flexibility and economics. Technologies such as weld overlay and chromizing are routinely applied by ALSTOM Power Inc. on the outside diameter (OD) surfaces to ensure that pressure parts are adequately protected from the environment in which they must operate. These techniques must be optimized for use in ultrasupercritical boilers. Diffusion coatings, utilizing the pack cementation method, may represent the best alternative for the protection of the inside diameter (ID) surfaces of tubing against the effect of high temperature oxidation.

In a coal-fired boiler, there are oxidizing and corrosive environments that range from simple gas attack to under-deposit microclimates of complex nature. The gases can be oxidizing, such as mixtures of O_2 and SO_2/SO_3, or a more complex mixture including aggressive gaseous compounds such as H_2S, HCl, COS, CS_2, CO and methyl mercaptan. These latter gaseous compounds may be generated during the substoichiometric combustion of coals when modified combustion systems are implemented for NO_x emissions control. Similarly, the substoichiometric combustion process generates unburned carbon and pyritic particulate that, based on the aerodynamics of the fireball, may end up deposited on heat transfer surfaces. The deposits can generate various local reducing environments ranging from carbonaceous to sulfidizing and even low-melting eutectics that act as a flux on the metal surface. The myriad of aggressive environments dictates a careful consideration regarding the quality of the surface-modified layers.

Boiler design considerations

Materials for USC coal-fired boilers will be selected on the basis of various criteria including:

- Mechanical Strength
- Ease of Fabrication
- Environmental Resistance
- Code Approval
- Cost

It is anticipated that over the range of USC boiler material conditions that there will be tradeoffs among these criteria. It is likely that there will be some materials that have satisfactory mechanical properties for use at a particular point in the cycle, but inadequate environmental resistance, either waterside or steamside. Consequently, a task has been initiated to examine the options for surface modifications that may be suitable to provide environmental resistance to lower cost alloys as an option to the utilization of more highly alloyed solid materials that are suitable for all aspects of service.

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A design study for an ultrasupercritical steam cycle was executed to assess feasibility of the use of advanced high-temperature alloys to construct a boiler (1). At the conclusion of this study, the conceptual design of a 700 MW steam generator for steam turbine throttle conditions of 5500 psi/1350°F/1400°F (38.5 MPa/730°C/760°C) was developed and materials required for high temperature pressure part components were defined. Figure 1 presents a side elevation of this ultrasupercritical boiler.

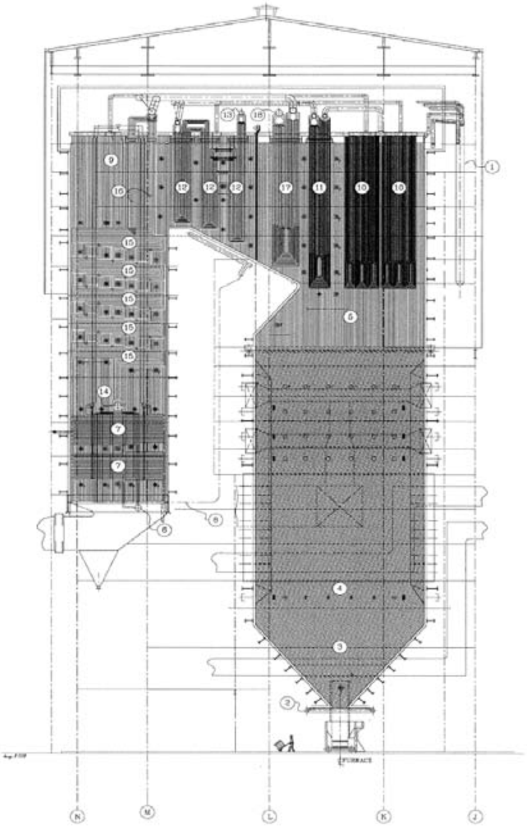

Figure 1: ALSTOM Power Inc. conceptual boiler design for 38.5MPa/730°C/760°C (5500psi/13500F/14000F).

Heat liberated in the furnace is partially absorbed by the waterwall tubes, forming the lower and upper chambers of the furnace. Combustion takes place in the lower furnace. The flue gas next enters the upper furnace where wide-spaced superheat division panels and superheat platens further cool the gases. Downstream of the platens and above the arch, there is a finishing reheater followed by a finishing superheater pendant. To minimize the effect of a high radiant heat flux emitted by the combustion process in the furnace, the final superheat section is shielded from the furnace and is installed behind the final reheat section. In the backpass, there is a primary reheater followed by an economizer. The economizer is the last section within the steam generator. The location of the convective and radiant surfaces is determined by considering a proper balance between gas, steam, and tube metal temperatures. Other surface arrangements, e.g. tower designs, could be employed also.

Metal temperature calculations were performed to establish material requirements. Material selections for the various boiler sections are summarized in Figure 2. The design includes waterwall fusion-welded panels. This design requires application of T23 (2Cr) alloy in the lower portion and T92 (9Cr) in the upper portion of the lower furnace. The upper, vertical wall section uses fusion-welded panels fabricated of T92.

The primary superheat duty is carried out in the roof and backpass wall tubes. Similar to the furnace walls, the likely candidate material of construction is T92. Next are superheat panels. Materials of construction and wall thickness vary depending on metal temperatures. Materials proposed are Super 304H, IN 617, and IN 740. The estimated maximum tube outside

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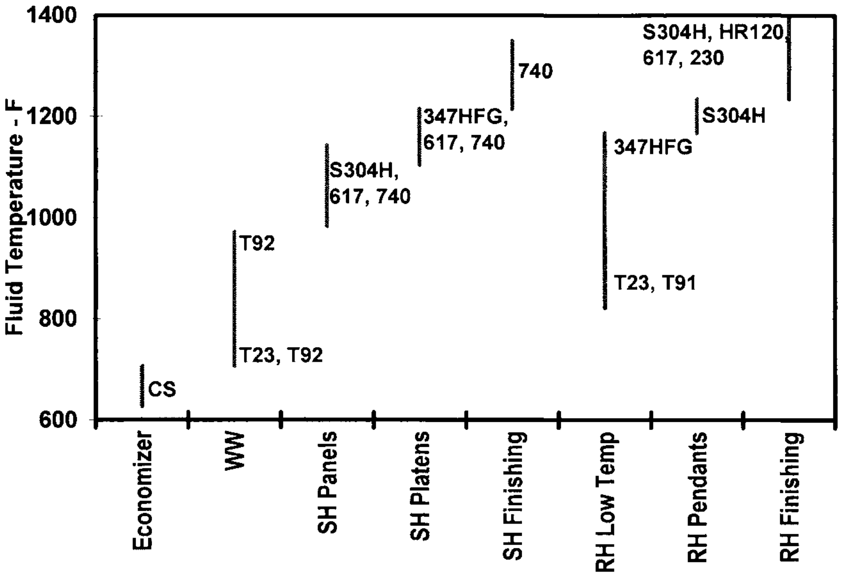

Figure 2: Material selections for conceptual ultrasupercritical boiler.

temperature can reach as high as 1330°F (720°C) in some parts of the panel, and the heaviest tube section has a minimum wall thickness of 0.42" (10.7mm). The superheat platens are next. As with the panels, materials of construction and wall thickness vary depending on metal temperatures. Materials considered for use are 347HFG, IN 617, and IN 740. The maximum tube outside temperature of about 1370°F (743°C) was calculated for the wrapper tubes (first tube in the assembly). The finishing superheat surface completes the heat transfer surface of the superheater. While IN 740 predominates in construction, IN 617 is applied for some sections of the tubes. Outside metal temperatures for some tube sections are above 1400°F (760°C). The heaviest minimum wall thickness used for IN 740 tubes is 0.200" (5.1mm).

Reheat duty is carried out in two sections: a primary reheat horizontal surface installed in the rear pass and a finishing surface located above the arch. Since the reheater operates at relatively low pressure, a range of materials are available with adequate strength to operate up to 1400°F (760°C), including Super 304H, HR 120, IN 617, and Haynes 230.

All of the materials noted above, with one exception, are ASME Code approved for boiler applications. The one exception is IN 740, a key alloy needed to achieve the target cycle steam temperatures, which is a developmental alloy. The nickel-based alloys, IN 617, IN 740, and Haynes 230 can be expected to cost as much as 8 – 10 times more per pound than the iron-based austenitic alloys. The high cost of the nickel-based alloys is compensated somewhat by their high strength and consequent lower wall thickness (i.e. lower weight). An economic study (2) of the proposed USC cycle concluded that improved cycle efficiency would support boiler and turbine capital cost of up to 50% more than current boilers at the economic breakeven point. Extending the range of usefulness of lower cost ferritic and austenitic materials via surface

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modification techniques will contribute to maximization of the economic value of the USC cycle.

Surface Modification Methods

This section of the paper will emphasize various surface modification techniques including thermal spray, claddings and diffusion coatings for the modification of heat transfer surfaces, both internal and external, for boiler applications. These techniques have recently been reviewed (3). Preliminary laboratory data will be presented to demonstrate the superior oxidation resistance of Cr-, Si-Cr-, and Al-Cr- diffusion coatings in superheated steam environments inside the boiler tubes.

Thermal Spray Techniques

Thermal spray coatings offer a wide range of material choices with high productivity in-field and in-shop applications, with low impact on tubing mechanical properties, and at a relatively low cost. Their performance depends on the control of specific parameters during application. The experience with thermal spray applications in the field, although limited for boilers, indicates that for long-term benefits, the applied coating needs to be virtually free of residual oxides and pores. It should also be free of residual stresses and preferably form a metallic bond to the substrate. Thermal sprays currently appear applicable only to the external tube surfaces.

There are three basic thermal spray quality parameters to consider for the surface modification of boiler heat-transfer components:

1. Bond strength: During the life of the boiler, structural components are subjected to fluctuation in load that can result in appreciable mechanical and thermal stress variations. Therefore, it is critical to the long-term integrity of the system that the coated layer has sufficient bond strength at the coating/base metal interface, with 8 ksi (55 MPa) being considered a minimum threshold value. A bond of metallurgical nature is preferable, although seldom attained with standard thermal spray methods. Therefore, an effective surface preparation prior to coating application is critical to generate the surface profile that will effectively "lock" the coating onto the surface.

2. Coating density-open porosity: Interconnecting porosity can undermine the integrity of the coating by allowing the permeation of aggressive gaseous species. The voluminous oxidation products formed at the interface at the expense of the base metal will ultimately lead to the exfoliation of the coating layer. Coating porosity should be below 0.5 % volume fraction.

3. Oxide content: Oxides are formed during the application process when the molten or semi-molten powder or wire has not been protected or shrouded with inert gases. Oxides present in the coating stack layers can lead to spallation under thermal cycling conditions. Oxide content should be below 1 %.

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The following thermal spray technologies satisfy these quality parameters and are proposed for boiler applications:

High Velocity OxyFuel (HVOF): The HVOF process produces coatings with relatively low levels of porosity and oxides (<1% volume fraction) due to high particle kinetic energy. This technique can generate coatings of quality similar to that of vacuum plasma spraying (high density and negligible permeability) at a lower cost. The coating adheres to the substrate by mechanical interlocking on the surface profile generated during grit blasting. The bond strength is of about 9 ksi (62 MPa) and 10.2 ksi (70 MPa) for ferrous and non-ferrous alloys, respectively.

Activated Combustion- High Velocity Air Fuel (AC-HVAF): This technique generates coatings that are virtually pore- and oxide- free (<0.10% volume fraction). The bond to the substrate is at least partially of the metallurgical type because of the intensive topochemical reactions at the substrate-particle interface. Due to high velocity impact, twice that of conventional HVOF, the projected particles undergo intensive plastic deformation to form a non-porous coating. The powder is sprayed below its melting point in an adjustable non-oxidizing combustion stoichiometry that results in minimal oxide inclusion. Once applied, the coating requires relatively low temperature input during unit start up to complete the sintering of the coated layer (4-6).

Cold Gas Dynamic Spray: Also known as High-Velocity Particle Consolidation (HVPC) (7), this process results in a dense coating with no porosity or oxide inclusions. The coating material in the form of powder is heated to only moderate temperatures, thereby minimizing powder oxidation. The particles attain sufficient kinetic energy to cause severe deformation upon impact, forming a quasi-metallurgical bond to the substrate surface. An inert carrier gas is used to accelerate the two-phase flow to supersonic velocities (300 –1000 m/s). This process is still in experimental design for large area application and further research is needed to understand fundamental process control methods.

These three processes have in common the ability of using powder. Powder utilization maximizes the choice of alloy chemistries to be applied, and opens the possibility of tailoring compositions with fewer restrictions as compared with monolithic alloys. The implementation into the market of nanocrystaline powders is providing coated layers with glassy characteristics. Amorphous layers are of interest since the lack of grain boundaries improves the oxidation/corrosion resistance of densified coating layers.

Claddings

Other surface modification techniques include weld overlay, laser cladding and composite tubing, which, like thermal sprays, are again applicable only to the tube external surfaces. Weld overlay techniques have been substantially improved and fully automated systems are now available. Distortion of the tubing is a concern, but in-shop applications minimize the problem by running water through the tubes during welding. Dilution of the weld wire typically is less

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than 10%, and it can be controlled by specifying filler metals with critical element concentrations at the high end of the compositional range.

During laser cladding, a laser-beam is used as the energy source to melt a pre-placed powder, a thermally sprayed layer or powder fed directly into the beam. Similar to weld overlay, the bond to the substrate is metallurgical in nature. Because of precise process control, laser cladding can generate a surface layer that approaches that of undiluted high alloy filler material. Laser cladding is currently limited to in-shop application, although the development of high power diode lasers may eventually allow for field applications.

Composite tubing has been used extensively in pulp-and-paper and petrochemical applications. For boiler applications, the use of an inner Ni-base alloy core may result in much lower oxidation rates than those experienced by conventional ferrous materials exposed to superheated steam. However, the nature of the aggressive environments generated in modified combustion units suggests that perhaps a heat transfer surface made of purely Ni-base metallurgies is required. The cost, however, can be prohibitive and the benefits of using Ni-base alloys solely on the ID is undermined by the lack of oxidation and/or corrosion resistance of conventional ferrous alloys exposed to fireside (or OD) conditions. It is more reasonable to approach the situation by modifying the surface of both ID and OD of ferrous alloy tubing by means of diffusion coatings.

Diffusion Coatings

Diffusion coatings generated by the pack-cementation technique can form layers rich in alloying elements that are beneficial for the myriad of aggressive environments that may exist in a coal-fired unit. In ferrous alloys, the diffusion layer is generated with minimal change of the original tubing dimensions. The attainable concentrations of chromium, silicon, and aluminum far exceed the maximum allowable concentration of conventionally manufactured alloys. The major drawback associated with multiple element coating is the lack of final composition prediction. Pack powders are tailored to generate compositions within acceptable ranges determined by the nature of the intended application. Diffusion coatings are commercially applied to tubing in both the power and petrochemical industries and are applicable to both internal and external tube surfaces.

Preliminary Test Results in Steam Oxidation

Steam is an oxidizing medium that reacts with metals to form an oxide layer. The oxide layer is generally beneficial in that it acts to seal off the metal surface from further oxidation. The oxide scale can be detrimental if it grows too thick since this provides additional thermal resistance and increases the tube wall temperature. The oxide scale can also be detrimental if it breaks off (exfoliates) from the tube surface and subsequently result in tube blockage or solid particle erosion in the turbine. Ferritic materials can be expected to oxidize in steam at the greatest rate, followed by stainless steel materials. The nickel-based alloys are the most oxidation resistant.

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Figure 3 shows the kinetics of oxidation of the Ni-base alloys 617 and Haynes 230 in superheated steam at 750 °C. Both alloys showed nearly parabolic behavior, indicating that the oxide layer is functioning in a protective manner to limit the oxide growth. The metallographic evaluation of the 617 test sample revealed a continuous oxide layer about 2-3 μm thick. Chemical analysis indicated the oxide to be chromium oxide with up to 5 % silicon. Protrusions were observed extending about 3 μm below the oxide layer. The oxide product in these indentations contained mainly Ni, Cr and Al. Cobalt and molybdenum were also detected. The Haynes 230 test samples were covered with a uniform chromium oxide layer about 1 μm thick. Some shallow oxide penetrations were observed.

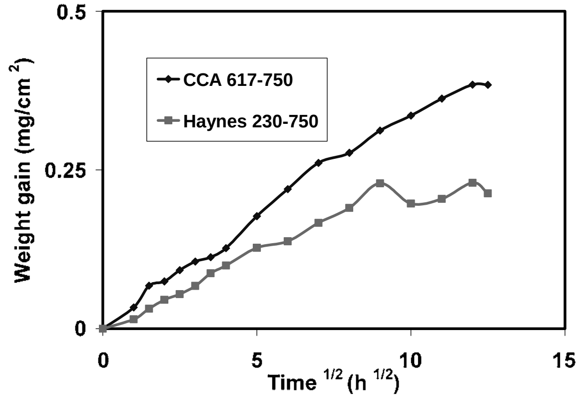

Figure 3: Oxidation of the Ni-base alloys 617 and Haynes 230 during exposure to superheated steam at 750 °C.

Figure 4 depicts the oxidation kinetics of diffusion coated T-92 samples in superheated steam at 750 °C. The coatings included a chromized layer, a Si-Cr rich layer and Al-Cr layer. All coated samples showed excellent oxidation behavior with the chromized sample having the least oxygen uptake. The metallographic evaluation did not reveal any evidence of oxidation. Figure 5 documents the condition of a chromized T-92 specimen after exposure to steam at 750°C for 160 hours. Figure 6 is presented for comparison to illustrate the growth of oxide scale on uncoated T-92 exposed at 650°C for a similar time period.

Figure 7 compares the kinetics of oxidation in superheated steam at 750 °C for the alloys 617, Haynes 230, Super 304H and the chromized T-92 specimen. The oxygen pick up for the alloys Super 304H and Haynes 230 are somewhat better than that of 617. The metallographic evaluation of the Super 304H specimen revealed a rather localized attack with the formation of iron oxides. The penetration was shallow and apparently followed grain boundaries. By comparison, the T-92 chromized sample shows a superior performance.

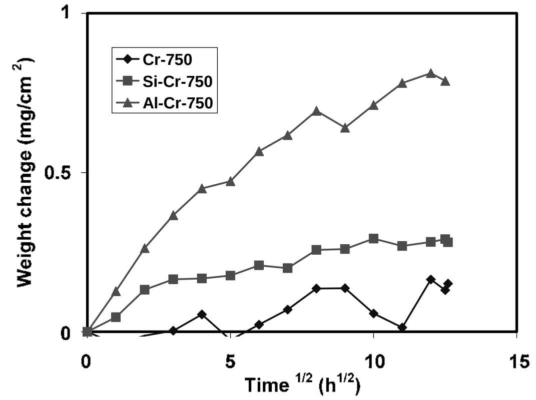

Figure 4: Oxidation kinetics for the diffusion layers in superheated steam at 750 °C.

Discussion of the steam oxidation performance of other surface modification techniques have

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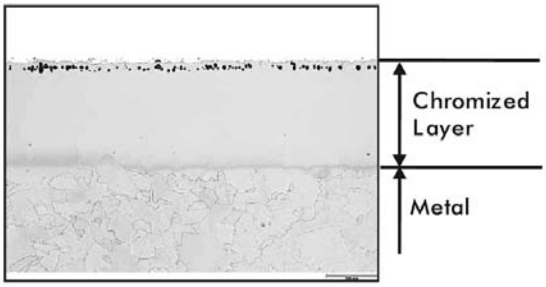

Figure 5: Chromized T-92 with no significant oxide layer present.

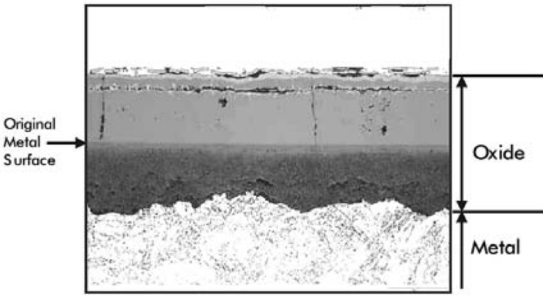

Figure 6: Oxide layer formed on uncoated T-92 at 650°C.

been reported (8). More extensive and longer term steam exposure testing is also being carried out in another portion of this program (9).

Economic Evaluation

Costs for the various alloys used to construct an ultrasupercritical, coal-fired boiler vary significantly from inexpensive low alloy steels to costly nickel-based alloys. Figure 8 provides an overview of the relative raw material cost of a number of alloys with the basis being the cost for low-alloy Grade 22. Differences in fabrication costs are not

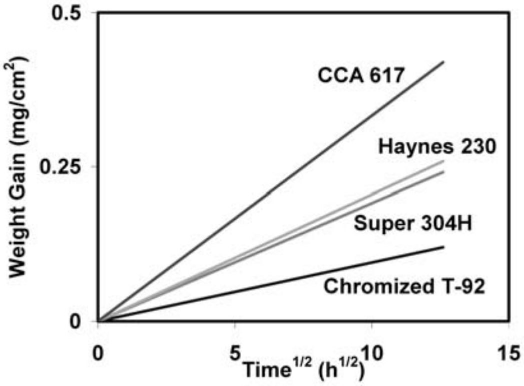

Figure 7: Comparison of oxidation rates.

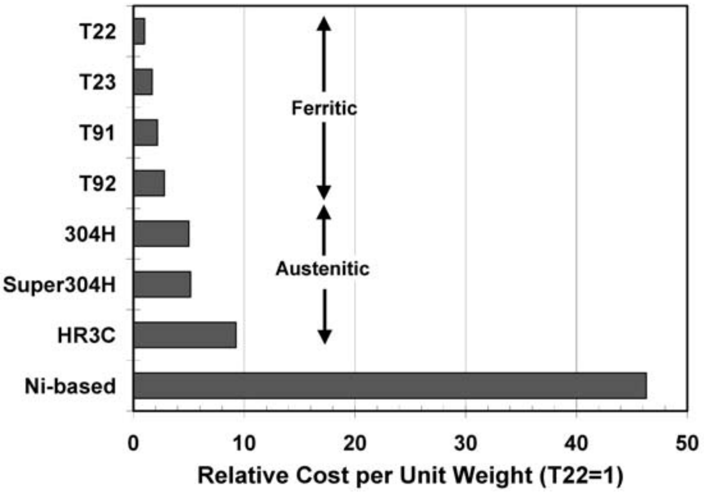

Figure 8: Relative cost of candidate boiler alloys.

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included and may ultimately affect the installed cost for use of a particular material.

It is apparent that boiler cost will be minimized by utilizing ferritic and austenitic alloys to their maximum capabilities of strength and environmental resistance. Figure 9 illustrates the strength limitations on service temperature based on the simplifying assumption that the tube wall thickness, calculated per ASME Code, will be no more than 25% of the tube outside diameter. Other boiler design restrictions may further limit the useful application temperature limit for a given material. The nickel-based alloys IN 740, Haynes 230, and IN 617 have a clear advantage in creep strength at the high temperatures employed in USC boilers.

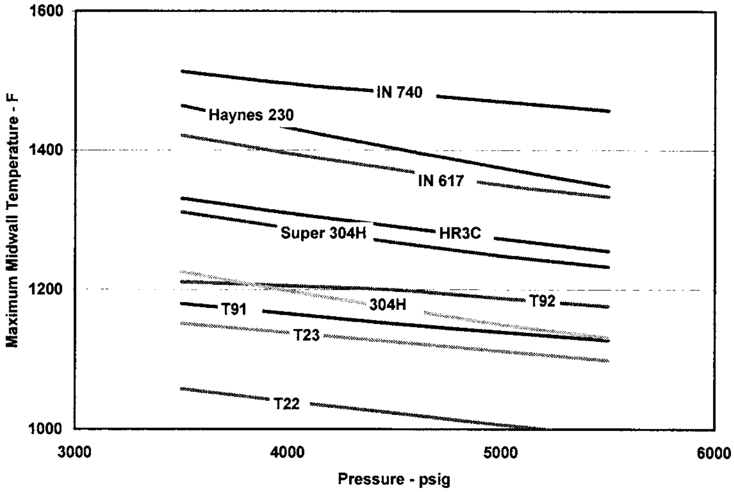

Figure 9: Service temperature limit based on strength for boiler tube materials.

The other aspect of materials service temperature limitation is the resistance to fireside and steam-side corrosion. An overview of the performance of various boiler materials relative to steam oxidation is shown in Figure 10. In this illustration, the calculated oxidation rate constant

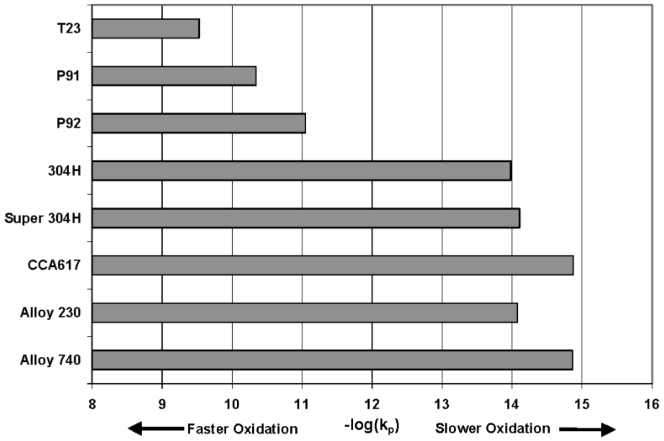

Figure 10: Steam oxidation rate constant for various boiler materials.

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(9) at 650°C, assuming a parabolic oxide growth model, k_p is shown for a selection of ferritic, austenitic, and nickel-based materials. It should be noted that while the austenitic materials have relatively low oxidation rates, they can be subject to undesirable exfoliation of the thin oxide scale that is formed. Thus, the austenitics may also benefit from surface modification.

As previously discussed, surface modification can improve the oxidation characteristics of both ferritic and austenitic substrates to achieve performance similar to the nickel-based alloys. Preliminary laboratory results are also indicating similar performance improvements relative to fireside corrosion, i.e. ferritic and austenitic alloy performance similar to the nickel alloys.

Surface modification treatments can be broadly expected to cost in the range of \$100 to \$200 per square foot (€1'300-€2'600 /m^2). An indication of the cost-savings possible for coatings is presented by two examples in Table 1, below. Assuming a generic coating cost of \$150 per square foot and the need for protection from the internal and/or external environment, superheater and reheater applications are estimated for various coating/substrate combinations.

Application	Superheater			Reheater	
Temperature	1200F (649C)			1400F (760C)	
Material Type	T92	Super304H	IN617	Super304H	IN617
Material Cost/ft	\$ 11	\$ 21	\$ 158	\$ 22	\$ 150
Coating Cost/ft	\$ 79	\$ 79	\$ -	\$ 98	\$ -
Total Cost/ft	\$ 90	\$ 100	\$ 158	\$ 120	\$ 150
Cost Ratio	57%	63%	100%	80%	100%

Table 1: Comparison of costs.

While more specific analysis of the costs and performance of particular substrate-surface modification method combinations is needed to solidify judgments on suitable applications, the cost benefits are apparent.

Conclusions

Ultrasupercritical boilers being considered for future, high-efficiency power stations will require the utilization of advanced, high-strength alloys. These alloys exhibit markedly improved mechanical properties compared with those typically used today, but are also very expensive. The boiler will expose these alloys to conditions where internal steam oxidation and external coal-ash corrosion can be expected to adversely affect service life. Surface modification methods are under review as a way to mitigate alloy degradation and permit the use of a less expensive alloy composition. Preliminary results to date, relative to steam oxidation, have shown the potential for coatings to significantly enhance long-term performance based on the suppression of oxidation effects. Specifically:

- The Ni-base alloys 617 and Haynes 230 exhibited adequate oxidation resistance in superheated steam at 750 C.

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- Super 304H alloy, although exhibiting low weight gains, experienced localized attack with the formation of Fe-oxides.
- The diffusion-coated T-92 alloy did not experience any extent of oxidation even though there was some oxygen uptake.
- Chromized T-92 offers superior resistance to oxidation in superheated steam.
- Preliminary economic assessment supports the use of surface-modified substrates up to their strength limits, rather than upgrading to more environmentally resistant alloys.

Improvement of steam oxidation performance by diffusion coating appears technically feasible and the best identified option at this time. The choices for surface modification techniques for coal-ash corrosion protection are broader. Continuing studies are expected to be able to expand these results through longer term testing and extension to fireside corrosion issues and ultimately to provide more specific economic justification. While the specifics of alloy selection will vary with boiler design details and the market being served, surface modification techniques are expected to provide additional options for the boiler designer.

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