Materials Characterization of New Composite Boiler Tube Materials

Final Report

to

Sumitomo Metals Industries
8750 W. Bryn Mawr Ave STE 1000
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by

Camden R. Hubbard[1], Gorti Sarma[2], Wally Porter[1], and Steve Spooner[3]
[1]Metals and Ceramics Division
[2]Computer Science and Mathematics Division
[3]Solid State Division
Oak Ridge National Laboratory
Oak Ridge, Tennessee

Summary of the Project Objectives and Tasks

ORNL facilities and expertise for neutron and x-ray residual stress analysis and thermal expansion were used to characterize the materials properties and boundary conditions (residual stress distribution) that are required to model how new candidate clad tubes will perform in service in a black liquor kraft recovery boiler. Sumitomo supplied two new alloys (HR11N and Super625) to be characterized in the form of 2.5" OD clad tubes and bar stock.

The measurement of thermal expansion as a function of temperature and residual stresses at the surface and through thickness will be used in ORNL computational models for boiler floor operation to predict the residual stresses as a function of temperature. The modeling data will indicate if tensile residual stresses, an essential factor in stress corrosion cracking, are present or not during any portion of boiler start up, operation or shutdown. The results will be compared to results published on 304L and 825 clad tubes.

The project had four tasks. Task 1 involved x-ray and neutron diffraction measurements of residual stresses at the surface and through the thickness of the clad tubes of both clad tubes. Determination of the linear thermal expansion of each of the alloys was the goal of Task 2. The results of both Task 1 and Task 2 were to be used in Task 3, the use of Finite Element Modeling to calculate changes in stresses in the composite tubes subjected to normal operating conditions of a black liquor recovery boiler. The fourth and final task involved presentation of an oral report and a preparation of a final letter report.

The power point presentation materials have been updated from those of the oral report presented on March 27, 2002 and are presented and referred to as figures in this final report. The figure number is in the lower right corner.

Thermal Expansion Measurements

Thermal expansion measurements for Super 625 and HR11N were measured using a Theta dual push rod dilatometer. A 25.0-mm long rod shaped specimen was prepared from the bar stock of each alloy. Measurements were obtained from room temperature (25°C) up to 1000°C and back to room temperature with a heating and cooling rate of 3°C per minute. Two full cycles of heating and cooling were recorded.

The linear expansion of HR11N is presented in Figure 5. Data for cycle 2 compared to cycle 1 show a small offset, likely due to relief of the cold work. Figure 6 presents the mean coefficient of thermal expansion (CTE) while Figure 7 presents the instantaneous coefficient of thermal expansion. The instantaneous CTE plot shows not only the relief of cold work but also a phase transition at between 550°C and 600°C.

The linear expansion data for Super 625 is presented in Figure 8, 9 and 10. Again, the cycle 1 and cycle 2 show a small offset (Figure 8) and the instantaneous coefficient of thermal expansion (Figure 10) shows both the change in length associated with annealing the cold work damage and the smaller expansion event likely associated with a phase transition.

There was a permanent change in length in both alloys after the testing related to the large downward peak in the iCTE curves for the first heat cycle. This is likely be an effect from the rolled condition of the plate or bar stock. The likely phase transition in both alloys is most likely related to the 590°C peritectoid reaction that occurs in the binary NiCr system. After the first cycle of heating the behavior seems to be reversible.

A comparison of the temperature dependence of the mean CTE with related materials - alloy 825, alloy 625, 304L stainless steel, and carbon steel SA210 is given in Figure 11. Super 625 has a higher CTE than either Alloy 625 and carbon steel SA210. However, the CTE of these there materials is lower than that of the other alloys. The CTE of HR11N and alloy 825 are very similar.

Residual Stress Measurements

A brief summary of the purpose and methods of x-ray and neutron diffraction for residual stress determination are provided in Figures 14 to 17. Figure 18 shows a portion of a boiler tube panel mounted for through thickness residual stress measurements by neutron diffraction methods at ORNL's HFIR. Figure 19 defines the principle directions for strain measurement used in ORNL's study of composite tubes. A summary of some of the results obtained at ORNL in prior studies on clad tubes is provided in Figures 20 to 22.

X-ray diffraction methods were used to measure the residual stress of the two Sumitomo clad tubes at a number of electropolished locations along the tubes. The depth of the electropolished spots was approximately 0.095 mm. Specific measurement details are provided in Figure 24. The residual stresses were found to be highly compressive (Figure 25), opposite to the prediction based on the CTE differences of the clad and SA210 of tensile stresses. Examination suggested a shot peening and Sumitomo confirmed this post straightening treatment. Straightening could also introduce a compressive surface residual stress. Stresses from straightening, however, have not been observed to be so large in other clad tubes. The depth of compressive stresses clearly must extend beyond 0.095 mm.

Neutron diffraction measurements were performed at the University of Missouri research reactor (Figure 26 shows the set up) as ORNL's High Flux Isotope Reactor was undergoing upgrades. The lower flux at UMRR and the limits of measurement time required a larger gage volume than in prior studies at ORNL. The results are shown in Figures 27 to 31. Note that the residual stresses for the mid point of the thickness of the clad layer for HR11N are neutral to slightly tensile while that for Super 625 are compressive. The through thickness stress results for HR11N/SA210 are much higher in the carbon steel near the interface than those measured for alloy 825. The smaller stresses in the Super 625/SA210 carbon steel clad tube are consistent with the small difference in CTE values compared to HR11N.

Finite Element Modeling of Stresses During Normal Operating Cycle

The X-ray and neutron diffraction measurements on the Super 625 and HR11N clad composite tubes were used to generate initial stress values through the tube thickness. These values were used in the modeling of the welding process to join the tube and the membrane using a two-dimensional finite element mesh. The resulting stress distribution was used to initialize the stresses for simulations of a normal operating cycle using a three-dimensional model of a floor tube panel. A more detailed description of the procedure may be found in a recent paper[1].

The analyses consisted of modeling the temperature distribution in the floor panel starting from room temperature through heating to operating conditions, followed by cooling back to room temperature. The resulting stress distribution was computed separately based on the temperature field. The initial stresses are altered by the difference in the coefficient of thermal expansion (CTE) between the clad material and the SA210 carbon steel, and are also influenced by the mechanical properties. Figures 33 and 34 show the variation of the coefficient of thermal expansion and the yield stress of the different materials with temperature, respectively. It is observed that Super 625 has a CTE value slightly higher than that of Alloy 625 and SA210 carbon steel, while the CTE of HR11N is very close to that of Alloy 825. The differences in yield stress are much larger, with Super 625 and HR11N having much lower values compared to Alloy 625 and Alloy 825, respectively. It should be mentioned that the property values for Super 625 and HR11N were based on data provided by Sumitomo, while data for other materials were based on values compiled from various handbooks on material properties.

For 304L stainless steel (304LSS) clad tubes, the large mismatch in the thermal expansion causes large stress changes during heating and cooling, and the low yield stress of 304LSS causes plastic deformation due to the stress changes. Therefore, these tubes have tensile axial and hoop stresses at the crown of the tube on the fireside surface at the end of a normal operating cycle. This is illustrated in Figure 35, which shows the changes in hoop and axial stresses for different clad materials, as the temperature changes from room temperature to operating temperature, and back again to room temperature. The initial compressive surface stress values for Super 625 and HR11N are based on x-ray diffraction measurements. It must be noted that the stress values are shown only at the final temperatures and are joined by straight lines, and the detailed variation as the temperature changes is not shown here. For Alloy 825, the CTE is closer to that of SA210 compared to 304LSS, and hence the change in stress is not as large. For alloy 825 the deformation remains in the elastic range, and the stress values return to their initial levels at the end of the operating cycle. Alloy 625, with an even closer CTE to that of SA210, also shows the same behavior and the stresses remain in the elastic range.

For both the Super 625 and HR11N clad tubes, the initial axial stresses are highly compressive, apparently a consequence of the shot peening used on the tubes during manufacturing. The hoop stress values are tensile, but are much lower in magnitude compared to the other clad materials. The initial hoop stress values in the tube were not measured by X-ray diffraction. Therefore, an assumption was made that they were the same as the axial stress values. Upon modeling the welding of tube and membrane to make the panel, the axial stresses remained compressive, but the hoop stresses became slightly tensile. Under normal operating conditions, the stresses at the surface become more compressive (or less tensile) due to effect of the thermal expansion mismatch with the SA210 carbon steel. This is seen quite clearly in the hoop stress variation for the different materials. The axial stress also becomes even more compressive initially, but soon

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[1]G.B. Sarma, J.R. Keiser, X.-L. Wang and R.W. Swindeman, "Modeling Studies to Predict Stresses in Composite Floor Tubes of Black Liquor Recovery Boilers," *J. Eng. Mater. Technol.*, 123 (3), 2001, pp. 349-354.

there is yielding at the surface of the clad material for Super 625 and HR11N. This is because the yield stress decreases with temperature (see Figure 35), and further increase in temperature causes the material to deform plastically at a lower stress magnitude. When normal operating temperature is reached, the axial stress magnitude is actually lower than the initial value. Upon cooling, the trend is reversed, and the stress at the outer surface becomes less compressive (or more tensile). Both Super 625 and HR11N have undergone plastic deformation during heating, and hence the axial stress upon cooling to room temperature is much different from the initial value. The hoop stress is also altered, but the change in magnitude is quite small.

The initial stresses measured at the surface of the tube using X-ray diffraction were quite high, and due to the large gauge volume for the neutron diffraction measurements, only one measurement was possible approximately at mid-thickness in the clad layer, and this value was much lower than the surface measurement. Due to the large difference in the values from X-ray and neutron diffraction in the clad layer, it was considered worthwhile to try another simulation where the surface value was made the same as the value at mid-thickness. The simulation for welding the tube and membrane was repeated, followed by analysis of a normal operating cycle, and Figure 36 shows the results for this second case. The stress values for the Super 625 clad tube are not significantly affected, but the values for the HR11N clad tube are now quite different. Both hoop and axial stresses for HR11N are now closer to Alloy 825. The change in stress is also comparable, due to the similar CTE values for both these materials. However, the lower yield stress for HR11N still allows plastic deformation, and hence the overall trend is similar to the previous case and the stresses do not return to their initial values, especially for the axial direction.

Even when the stress values at the surface of the clad layer were based on extrapolating the neutron diffraction measurement at the mid-thickness to the surface, the stresses for Super 625 in the panel were quite high in compression. Therefore, a hypothetical case was considered next, where the initial stress values for the panel were assigned based on values calculated earlier for the Alloy 625 clad tubes and membranes. A similar approach for HR11N clad material was adopted based on Alloy 825. The resulting stress variation is shown in Figure 37, allowing for a more direct comparison between Super 625 and Alloy 625, and between HR11N and Alloy 825. As seen from Figure 37, the initial stress values are about the same for the corresponding materials. However, the lower yield stress values for Super 625 and HR11N play a role in causing plastic deformation during the operating cycle, while stresses in Alloy 625 and Alloy 825 remain in the elastic range. Hence, the basic trend in the behavior of these materials is not altered even when the initial stresses are made similar to the materials studied earlier.

• Summary

A comparison of the different clad materials indicates that 304LSS undergoes the largest changes in stress values due to the largest mismatch in CTE with that of SA210 and the smallest yield stress. The other materials show comparatively smaller changes in stress, and their behavior is qualitatively similar, although there are considerable differences in the stress magnitudes. It is commonly observed that cracks on floor tubes in recovery boilers are mainly circumferential, indicating that tensile axial stresses play a role in their initiation and propagation. The axial stress values after a normal operating cycle for clad materials other than 304LSS are mostly compressive to varying degrees, the only exception being the HR11N clad tube for the last case, where the initial stress is based on the values calculated earlier for Alloy 825. The higher yield properties for Alloy 825 and Alloy 625 lead to stress behavior that remains elastic, so that stress remain at initial levels, whereas comparatively lower yield stress causes plastic deformation and change in axial stress for Super 625 and HR11N. However, once the first cycle is completed, the axial stress magnitudes are not as high for these materials. Therefore, additional cycles would not be expected to cause further plastic deformation. This has been verified through modeling, and Figure 38 shows the results for Super 625 and HR11N clad tubes over two cycles of normal operation. The initial surface stresses in the tube for the case shown in Figure 38 were based on the X-ray diffraction measurements, but other cases also gave similar results, with the second cycle causing no further plastic deformation.

• Limitations of modeling

Several simplifying assumptions have been used to model the stress in the floor tube panels, and the predictions are also dependent on the material properties used. Therefore, it is important to place greater emphasis on the trends in the stress variations, such as presence of tensile axial stress in 304LSS, and compressive axial stress in other materials, rather than on the exact values generated by the current modeling. The differences between Super 625 and Alloy 625, and between HR11N and Alloy 825, may be larger than might have been expected based on the similar compositions of these materials. The differences are enhanced by the higher yield properties of Alloy 625 and Alloy 825 used in the modeling, but materials with these properties may have been intended for uses other than composite tubes in recovery boilers.

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