Modeling the Ferrite-to-Austenite Transformation in the Heat-Affected Zone of Stainless Steel Multi-pass Welds

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Abstract

The ferrite/austenite transformation in austenitic stainless steel welds was modeled by considering the transformation as a diffusion-controlled process. A finite-difference method was used to solve the diffusion equations. The transformation behavior was evaluated for both isothermal aging and linear cooling over a range of cooling rates. The analysis provides information on the change in ferrite content as a function of heat treatment as well as the rate of the formation (or dissolution) of ferrite. The compositions of the ferrite and austenite are also obtained in the analysis. The results show that the approach to equilibrium can often be indirect and sometimes counter to intuition. The analysis is useful in providing information on the microstructural stability in austenitic stainless steel welds that cannot be obtained experimentally.

KNOWLEDGE OF THE MATERIAL MICROSTRUCTURE is critical in order to understand and project how a material will respond to further processing and to service conditions. The microstructure controls the properties such as strength, corrosion resistance, toughness, ductility, and weldability. In addition, a thorough understanding of the microstructure is important in evaluating the alloy stability when exposed to elevated temperatures during service. This is particularly important in welding where the heat-affected zone (HAZ) may be subjected to temperatures up to the solidus temperature. Finally, in the case of welds, the microstructure in the HAZ may be very important because this region acts as the "mold" for solidification of the weld. Due to the epitaxial nature of solidification during welding, the microstructure of the HAZ at the time of solidification may have a significant influence on the solidification process and the final weld microstructure.

There are several means by which the microstructure can be assessed. Direct observation is the most straightforward approach, but it is limited by the extent to which the microstructures and phases present in the HAZ at elevated

temperatures can be preserved at the observation temperature. Equilibrium phase diagrams can also provide much guidance in evaluating the microstructure. In the case of weldments, constitution diagrams such as the Schaeffler diagram (1) and recent revisions (2) provide additional information on the weld microstructure. However, these diagrams also have limitations. Equilibrium diagrams are of limited value because equilibrium is often not achieved under normal welding conditions. Constitution diagrams cover a wide range of alloy compositions by invoking nickel and chromium equivalents. As such, they are a very valuable first approximation, but they may be of limited accuracy for specific alloys since they represent average behavior. In addition, constitution diagrams do not consider all variables that may have an influence on the microstructure, such as cooling rate, and therefore their predictions may not be accurate under certain conditions (3).

For welding conditions, the material in the HAZ is subjected to very high temperatures. At these temperatures, experimental evaluation of the phase stability and transformation behavior is extremely difficult, if not impossible. In addition, experimental studies are alloy specific and therefore cannot be automatically expanded to other alloy compositions. Because of these limitations, modeling of the microstructural development is advantageous and desirable. Modeling can be readily applied to different compositions, and can provide critical information on the high-temperature phase stability that cannot be easily obtained otherwise. Modeling can also provide a wide variety of information, such as phase compositions, kinetics of the transformation, the influence of changing temperature cycles, and so on.

For austenitic stainless steel welds, the microstructure typically consists of two phases: an austenitic matrix phase (γ) with residual ferrite (α). Figure 1 shows a typical fusion zone (FZ) microstructure, with a ferrite content of approximately 10%. When exposed to elevated temperatures, either during subsequent welding passes or in service, the microstructure is not stable. Depending upon the exact thermal conditions, the ferrite may grow at high temperatures, or it may dissolve or transform at lower temperatures. Evidence of this phase instability is revealed in

Fig. 1 - Typical austenite plus ferrite microstructure in an austenitic stainless steel weld (type 304 S.S.).

Figure 1. The base material is fully austenitic, but in the HAZ, immediately adjacent to the FZ, some intergranular ferrite can be found, as indicated by the arrow. This ferrite must have formed during the exposure of this region to elevated temperatures during the thermal cycle. Direct observation, as in Figure 1, reveals that ferrite formed, but it does not give any indication as to the extent of ferrite formation at elevated temperatures, prior to cooling, and the extent of the dissolution of ferrite during cooling. If the ferrite formation is extensive at elevated temperatures, then its presence may have a profound effect on the solidification behavior. These issues cannot be readily resolved by direct observation, but modeling of the ferrite-austenite transformation can provide valuable insight into the rate and extent of the reaction at the elevated temperatures. In the present study, the ferrite-austenite transformation has been modeled by evaluating it as a diffusion-controlled transformation. Information such as the extent of ferrite formation, the ferrite composition, and the influence of cooling rate on these quantities can be readily provided by such a modeling approach.

Calculation Procedure

The model was based on the evaluation of the diffusion-controlled growth or dissolution of ferrite in an austenitic matrix. Nucleation of ferrite was not considered since the starting microstructure contained both phases. Figure 2(a) illustrates the geometry of the system that was modeled, and Figure 2(b) shows how this model system can be used to represent a standard volume element in the actual microstructure. Referring to Figure 2(a), a one-dimensional system of finite size L was considered, with a planar interface between the ferrite (size ℓ) and austenite (size $L - \ell$). In weldments, the ferrite exists at the dendrite cores, and it would be more realistic to assume a cylindrical geometry for the ferrite. However, it has been shown (4) that there is basically no difference in the kinetics of the transformation for planar, cylindrical, or spherical geometries as long as the ferrite is centered at the core of the system, as is appropriate for the modeling of austenitic stainless steels.

Fig. 2 - (a) Schematic of modeled system and (b) volume element, V , that can be used to model the entire microstructure.

For simplicity, the initial ferrite and austenite compositions were assumed to be constant (but different). Commercial stainless steel alloy compositions were modeled by considering an Fe-Cr-Ni ternary alloy. As will be shown, the use of ternary alloys leads to many interesting results that are typical of multicomponent systems in general, and these effects would not be observed if only a binary system was considered. This is because in binary systems, there is one tie-line in a two-phase field at each temperature. However, in ternary and higher order systems, the added degree of freedom allows for a range of tie-lines between ferrite and austenite at one temperature, and this added flexibility permits for many new types of behavior.

The analysis involved the solution of Fick's Second Law, given as:

$$\frac{\partial C}{\partial t} = D \frac{\partial^2 C}{\partial x^2} \quad (1)$$

where C represents the composition, t is the time, D is the diffusion coefficient, and x is the distance. At the interface between the ferrite and austenite phases, a mass balance is specified. This mass balance requires that the rate of growth of the ferrite is balanced by the net flux of chromium or nickel at the interface. Mathematically, this mass balance is given by:

$$-D_\alpha \frac{\partial C_\alpha}{\partial x} + D_\gamma \frac{\partial C_\gamma}{\partial x} = \frac{\partial x}{\partial t} (C_\gamma - C_\alpha) \quad (2)$$

The subscripts α and γ refer to the ferrite and austenite phases, respectively, and the concentration derivatives in the above

equation are evaluated at the ferrite/austenite interface. This mass balance must be satisfied for both the chromium and nickel constituents. The equilibrium interface compositions (tie-line compositions) are adjusted so that the interface velocities determined from the nickel and chromium mass balances are equal. An improved iteration scheme was utilized so that the mass balances were satisfied with a consistent set of compositions (4).

The diffusion equations were transformed into an implicit, finite-difference form following the approach described in the literature (5-7). The use of a finite-difference approach was required because of the combination of a ternary system, rather than a binary one, and the finite size of the total system. This latter condition allows for overlapping profiles from neighboring volume elements [see Figure 2(b)]. Without the finite size and the ternary system features, the diffusion equations could be solved by analytical means. A fixed number of nodes was specified for both the ferrite and austenite phases. The Murray-Landis transformation (7,8) was used to account for the interface movement when specifying the positions of the nodes. A similar approach has been used recently by Kikuchi, Kajihara, and co-workers (9-11) to study the ferrite-austenite transformation in stainless steel castings during isothermal aging at 1100°C. The present study concentrates on austenitic stainless steel compositions that are typical of welding alloys, and the calculations were carried out over a range of isothermal aging temperatures from 700 to 1300°C. In addition, the present study includes calculations for non-isothermal conditions in which a range of constant cooling rates were considered.

In the current analysis, the assumptions and simplifications included (1) composition-independent diffusion coefficients, (2) equal densities for ferrite and austenite (so that compositions could be expressed in weight percent), (3) cross-term diffusion coefficients were negligible, (4) the presence of sigma phase was ignored by not allowing it to form, and (5) equilibrium at the ferrite-austenite interface. The diffusion coefficients were temperature dependent. The equilibrium at the interface between ferrite and austenite was specified by the ThermoCalc™ program (12). The data from ThermoCalc provided the equilibrium ferrite and austenite compositions as a function of both temperature and overall alloy composition. These results were then incorporated into the finite-difference software code in an analytical form. The details of this procedure, and the actual analytical expressions that were used, are given elsewhere (7). It was shown that this analytical representation of the phase boundaries and tie-lines was quite accurate over the temperature range considered in this study, namely 700 to 1300°C. The mathematical expressions for the diffusion equations in finite-difference form, the diffusion parameters, and the accuracy of the methodology are described in detail in another paper (7).

The present paper describes results for two alloy compositions that are representative of austenitic weld filler metal compositions (in wt %): Fe-21.9Cr-10.35Ni and Fe-21Cr-11.3Ni. The starting compositions of the ferrite and austenite were Fe-30Cr-4.5Ni and Fe-21Cr-11Ni, respectively, for the first alloy, and Fe-30Cr-5Ni and Fe-20Cr-12Ni, respectively, for the second alloy. These ferrite and austenite compositions are typical for these phases in the as-welded condition, as determined by

analytical electron microscopy (13-15). The size of the system, L , was fixed at 5 μm while the initial ferrite size was 0.5 μm . As can be seen in Figure 2(b), these sizes are appropriate for a representative sample volume in the as-welded condition. The calculations were carried out for isothermal aging at 100° intervals from 700 to 1300°C until final equilibrium was reached at each temperature. Representative results for 1000 and 1200°C aging will be presented here; results for other temperatures may be found elsewhere (7). Calculations were also made for non-isothermal conditions, in which the system was cooled at a constant rate (0.1, 1, 10, or 100°C/s).

Results and Discussion

The finite-difference routine calculates the composition of ferrite and austenite as a function of distance and time. The location of the ferrite interface is also determined so that the ferrite growth (or dissolution) can be readily followed as a function of time as well. Figure 3 shows a plot of the composition profiles as a function of distance for selected isothermal aging times at 1000°C for the Fe-21.9Cr-10.35Ni alloy. The calculations show that the ferrite grows slightly while becoming slightly leaner in chromium and richer in nickel. Under these conditions the austenite composition changes only slightly. The ferrite size can be readily converted into a ferrite content. The change in ferrite content can be followed more easily by considering it as a function of aging time, as is done in Figure 4 for both alloys. The results for aging at 1000 and 1200°C are shown in Figures 4(a) and (b), respectively. Similarly, the average chromium and nickel compositions of ferrite and austenite as a function of aging time at

Fig. 3 - Typical calculated composition profiles for (a) chromium and (b) nickel after aging at 1000°C for selected times.

1000 and 1200°C are shown in Figures 5 and 6.

Several interesting features are found in Figures 4 through 6. First, for one alloy (Fe-21Cr-11.3Ni), aging at 1000°C results in the total dissolution of the ferrite, yielding a fully austenitic structure [see Figure 4(a)]. For computational reasons, less than 0.2% ferrite was designated as "totally dissolved." Thus, the model predicts the aging time required for complete homogenization and elimination of any residual ferrite. At 1200°C, the ferrite remains stable in this alloy. For the other alloy, the ferrite remains a stable constituent at all temperatures. In fact, at 1200°C, for both alloys, ferrite growth is observed. For the Fe-21.9Cr-10.35Ni alloy, the ferrite content increases to nearly 35%, and this growth is completed in less than 10,000 s. Even within 10 s, which is comparable to the exposure time of the HAZ during multipass welding, the ferrite content increases to more than 20%. At 1300°C, the kinetics are faster and the final amount of ferrite is even greater (7). These results indicate that the HAZ undergoes substantial microstructural changes during multipass welding, and these changes are likely to have an impact on the solidification behavior, final microstructure, and properties.

The figures also show that the approach to final equilibrium is often not direct. For example, for the Fe-21.9Cr-10.35Ni alloy at 1000°C, the ferrite chromium level initially decreases with aging, but with additional aging time, the chromium level recovers somewhat. This behavior was found under other conditions as well (7). It can also be seen that the change in ferrite content proceeds in two stages, as shown in Figure 4(b) for the Fe-21.9Cr-10.35Ni alloy. At other aging temperatures, it was

Fig. 4 - Calculated ferrite content versus aging time at (a) 1000°C and (b) 1200°C.

Fig. 5 - Average chromium compositions of ferrite and austenite as a function of aging time at (a) 1000°C and (b) 1200°C.

Fig. 6 - Average nickel compositions of ferrite and austenite as a function of aging time at (a) 1000°C and (b) 1200°C.

found that the ferrite content may increase before eventually decreasing, or vice versa (4,7,16). These indirect paths toward final equilibrium were investigated further, and it was determined that the effects were real; these were not artifacts of the calculation procedure. They arise from the condition of equilibrium at the interface and the required balance of chromium and nickel fluxes at the interface to yield a consistent value for the instantaneous growth rate of ferrite (4,7,16). Generally speaking, the behavior at early times is controlled by the ferrite diffusion, which is nearly two orders of magnitude greater than the diffusion in the austenite. At later times, the austenite diffusion controls the equilibration process. Due to the nature of ternary and higher component systems, the tie line that relates the ferrite and austenite compositions in equilibrium at the interface is not fixed. Thus, the equilibrium interface compositions may, and do, change as a function of time. As a consequence, the fluxes at the interface also change, and this may result in irregular behavior such as ferrite growth initially, followed by ferrite dissolution. The reversal from growth to dissolution has been observed experimentally in austenitic stainless steel diffusion couples (10). Similarly, the change in tie-line compositions can also lead to reversals in the ferrite or austenite compositions as a function of time. This behavior is examined in more detail elsewhere (7).

Calculations were also made for non-isothermal heat treatments in which different constant cooling rates were utilized to simulate the cooling of the HAZ during welding. These calculations were performed for the Fe-21Cr-11.3Ni alloy. The initial ferrite (23.6Cr, 8.58Ni) and austenite (19.49Cr, 12.88Ni) compositions, as well as the amount of ferrite (36.7%), were adjusted to represent approximately the compositions and amounts present in equilibrium at elevated temperatures (1300°C). The ThermoCalc program was utilized to determine the high-temperature equilibrium state. The procedure calculated the composition profiles in the ferrite and austenite during cooling from 1300 to 700°C, at a constant rate of 0.1, 1, 10, or 100°C/s. The ferrite content, and ferrite and austenite compositions are plotted as a function of temperature in Figures 7 through 9, respectively. The equilibrium ferrite content and ferrite and austenite compositions as a function of temperature are also shown in the figures (dashed lines are interpolations over temperature ranges where the ferrite is not stable). The plots show that, in

Fig. 7 - Calculated ferrite content versus temperature as a function of cooling rate from 1300 to 700°C.

general, as the cooling rate increases, the deviation from equilibrium increases as well. In addition, the curves show at what point the ferrite content, or phase compositions, no longer change appreciably with further cooling, in effect representing the temperature at which the microstructure is effectively quenched. As is to be expected, at the higher cooling rates, the compositions or ferrite content are quenched-in more quickly. These curves can be used to determine the residual ferrite content and the phase compositions that are retained at room temperature as a function of those process conditions which affect the cooling rate, such as power, plate size, and welding speed.

A curious effect is found for the ferrite nickel content as a function of cooling rate [see Figure 8(b)]. Whereas at the highest cooling rate (100°C/s), the ferrite nickel content deviates almost immediately from the equilibrium value and remains higher than the equilibrium compositions, the same is not true at the lower cooling rates. In fact, in the range from 1000 to 1200°C, the 10°C/s condition actually has a ferrite nickel content that is closer to equilibrium than that found at the lower cooling rates of 1 and 0.1°C/s. Furthermore, for the three lowest cooling rates, the ferrite nickel content is less than that found in equilibrium, indicating that the change in composition is actually greater than that for the equilibrium state. A more detailed examination of this behavior shows that it is a real effect that can be readily explained. As noted above, for some isothermal aging conditions, the ferrite nickel content may quickly decrease, initially, and then increase with additional aging time. This is, in fact, the situation for the Fe-21Cr-11.3Ni alloy at 1200°C, when the starting compositions are

Fig. 8 - Ferrite (a) chromium and (b) nickel compositions as a function of temperature for different cooling rates.

Fig. 9 - Austenite (a) chromium and (b) nickel compositions as a function of temperature for different cooling rates.

set at the equilibrium values at 1300°C. The starting ferrite nickel content is 8.58%, and the nickel concentration decreases quickly at first and then increases (4). During cooling at the slowest rates (0.1 and 1°C/s), there is sufficient time for the ferrite nickel level to reach this minimum value, while at the 10°C/s rate and higher, this minimum nickel level is not reached. Thus, the change in nickel content is less in the 10°C/s case than for slower cooling rates, and this smaller change is actually closer to the final equilibrium nickel level. It is noteworthy that this trend does not hold true for the chromium concentrations. Thus, the model calculations yield results that are understandable but not always intuitive. These findings suggest that model calculations are quite valuable, in that they can predict behavior that cannot be followed experimentally and would not be expected from simple intuitive arguments.

Summary

An implicit, finite-difference model has been developed and applied to study the diffusion-controlled ferrite-austenite transformation in austenitic stainless steel welds. The model was applied to two Fe-Cr-Ni alloy compositions. The calculations provide valuable information on the microstructural development in these steels as a function of thermal treatment. In particular, the conditions of isothermal aging at 700 to 1300°C and constant cooling from 1300°C were evaluated, and the quantities that were determined include ferrite content and average ferrite and austenite

compositions. The model results also provide valuable information on the kinetics of the transformation reactions. These quantities are desirable to know because they play a major role in determining the weld properties and may also be influential in the solidification process. Furthermore, these quantities are often difficult to measure experimentally, especially at the elevated temperatures to which the HAZ microstructures are exposed.

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