

FUNCTIONAL FORMS OF EQUATIONS TO PREDICT STEEL WELD METAL PROPERTIES

by

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ABSTRACT

Empirically derived expressions are commonly used to predict specific steel weldment properties. These expressions usually consider only constitutional considerations and are limited to their ability to predict the influence of the thermal experience (heat input, weld preparation and plate thickness). Carbon equivalent and basicity index are examples of such predictive expressions. This paper reviews some of the existing expressional forms and introduces new functional forms that are based on metallurgical engineering concepts. Forms for equations which can predict weld metal properties and behavior as a function of composition and cooling rate are suggested. The concept of developing isoproperty diagrams that allow better selection of welding consumables with variations in heat input is introduced and discussed.

INTRODUCTION

Historically, property predictive equations have been applied to wrought materials, and empirical expressions have been used to describe the influence of alloy additions on the austenitic decomposition of steels and to predict final transformation microstructures and properties. These equations play a very important quality control role in the fabrication of marine structures. Their use assures that the materials being purchased will be less susceptible to hydrogen cracking in the heat affected zone (HAZ). The newer thermomechanical control-rolled processes (TMCP) have produced steels with much higher HAZ toughness properties which are less susceptible to hydrogen cracking. Despite that the industry remains vigilant about the HAZ properties, there is increasing concern that the weld metal has become the weak link in the weldment with lower toughness values and more susceptible to hydrogen cracking. Extensive welding procedure qualifications are made to qualify these weld metals before fabrication, but predictive equations are not applied when specifying weld metals. More recently, however, attempts have been made to apply some of the predictive equations to weld metal microstructures.

The carbon equivalent (CE) equation is an example of such predictive expressions used to quantitatively decide the need of pre- and postweld heat treatment. Basicity index is another example, often used to predict weld metal oxygen content and mechanical properties.

Carbon Equivalent

Carbon equivalent equations are hardenability type expressions which were originally developed for use with medium carbon steels where eutectoid decomposition is the primary phase transformation with martensite as the major product during fast cooling. They are empirically determined to explain the influence of alloying elements on the transformation behavior of steels. They have also been successfully used for predicting weldability of various types of parent metal plates by correlating alloy content and cracking susceptibility during welding. A commonly used carbon equivalent equation is given below to illustrate weldability prediction (1).

$$CE = C + \frac{Mn}{6} + \frac{Cr - Mo + V}{5} + \frac{Ni + Cu}{15} \quad \dots\dots (1)$$

Alloys with carbon equivalent greater than 0.45 weight percent are usually welded with a preheat.

When carbon equivalent is greater than 0.55 weight percent, both pre- and postweld heat treatment are recommended to obtain a sound weld. Another carbon equivalent type expression that relates weld cracking susceptibility to alloy composition is the Ito-Bessyo P_{cm} equation (1.2).

$$P_{cm} = C + \frac{Si}{30} + \frac{Mn + Cu + Cr}{20} + \frac{Ni}{60} + \frac{Mo}{15} + \frac{V}{10} + 5B \quad \dots\dots (2)$$

The P_{cm} equation correlates well with the steels that have low carbon usually < 0.1 weight percent. Steels with P_{cm} values of 0.2 weight percent or lower have shown good resistance to weld cracking.

Even though the above equations are commonly applied in wrought materials it is questionable that these expressions have the ability to predict properties and transformation behavior of weld metals. Plate martensite of high carbon content is not expected to predominate in the weld metal microstructure and microalloy precipitates may form during the welding thermal cycle. Additionally, influences from solidification and second phase particles such as inclusions will also affect the weld metal solid state transformation reactions. Oxide inclusions are known to promote ferrite nucleation and should reduce the hardenability. The existing carbon equivalent expressions do not sufficiently introduce the role of oxide weld metal inclusions into the formulation, which needs to be done before the carbon equivalent equations can become reliably used for prediction of weld metal microstructure and properties.

Historical Development of Carbon Equivalent Type Equations

Grossmann (3) in 1942 proposed a method to determine hardenability of steels as a function of their chemical compositions and prior austenite grain sizes. He introduced the concept of Ideal Critical Diameter (DCI) which is expressed mathematically as :

$$DCI = D_{Base} \pi F \quad \dots\dots\dots (3)$$

The calculated ideal critical diameter, DCI relates

to the diameter of a cylindrical specimen which when water quenched results in a microstructure of 50 percent martensite at its centre. For each additional element in a "pure" iron alloy, a multiplication factor (F_i) was assigned, according to its weight percent in the alloy, and multiplied to the base hardenability. D_{Base} .

In leaner alloys, especially the high strength low alloy (HSLA) steels, there is difficulty in achieving fully martensitic structures. The method of relating martensite to hardenability, such as the Grossmann procedure, may not always be applicable to those alloys. Therefore, other analytical procedures are needed to determine the hardenability of the shallow hardening steels. As an example, Grange (4) in 1973 described the influence of alloying elements on hardenability as additive rather than multiplicative. The criterion used was ninety percent martensite in a hot brine quench test. In this case, the mathematical expression of the hardenability diameter (D_H) is

$$D_H = \sum_{i=1} \Delta D_x \quad \dots\dots (4)$$

where ΔD_x are the additive terms corresponding to the increase in hardenability due to the presence of each one of the alloying elements (X_i). It is assumed that the hardenable diameter (D_H) for iron is zero. Notice that Equation 4 is similar to the carbon equivalent and P_{cm} expressions in Equations 1 and 2. However, in order to modify the carbon equivalent equations, which were determined for wrought materials, such that they become effective in predicting weld metal properties, two major factors need to be added. They are : (i) the role of weld metal oxygen and (ii) the influence of welding parameters.

Role of Weld Metal Oxygen

Weld metal inclusions have been found to be an important factor in influencing weld metal microstructure in low carbon steels and high strength steels (5-10). The type, size distribution, and density of weld metal inclusions are all important variables to control and achieve the desired weld

metal microstructures and properties. Liu and Olson (10,11) suggested that for low carbon microalloyed steel weld metals, the desirable size distribution of inclusions would be when the maximum in size distribution is greater than the Zener particle diameter. This condition will promote larger prior austenite grains because inclusions of size larger than the Zener diameter will not hinder austenite grain growth. As a result, high toughness acicular ferrite will form intragranularly, nucleating on the inclusions. On the contrary, if large fraction of the inclusions are below the Zener diameter, a large number of inclusions will pin the austenite grain boundaries resulting in large fractions of grain boundary ferrite. Inclusions rich in titanium and aluminum were also found to nucleate acicular ferrite.

In high strength steel weld metals, Ramsay et al (12) found that a bimodal inclusion size distribution would be best to achieve the smallest austenite grain size and the smallest packets of lath martensite. This condition would allow optimum weld metal toughness at a given strength level. A fundamental understanding of weld pool chemistry is essential if the proper inclusion type, size distribution and quantity are going to be produced consistently over a specific heat input range.

Weld metal predictive equations must account for the weld metal oxygen and inclusion content. An oxygen term or terms need to be added to the carbon equivalent expressions and the magnitude of the coefficient is dependent on the type of inclusions being formed (the type of consumable being used). For low carbon steel weld metals, it is difficult to obtain reliable predictability from the carbon equivalent equation unless the complex oxygen and inclusion effect can be evaluated and incorporated. To date, the concept of basicity is often used to relate welding consumable composition to weld metal oxygen. A combination of both carbon equivalent and basicity should be used to understand weld metal transformation and properties. A similar concern must be addressed for weld metal nitrogen and sulfur.

Basicity Index

Basicity index is a formulation used to relate weld metal oxygen content and steel weld metal properties (13). The identification of high weld metal toughness in steel weld metal with the use of high basicity welding consumable is a common practice. However, the concept of slag basicity was originally developed as a sulfur index to describe the sulfur removal ability of a slag in steel making. As such, basicity was defined as a ratio with the strong sulfide and sulfate formers present in the numerator and the weak sulfide formers in the denominator. The mathematical expression of basicity index (BI) is shown in the equation below :

$$BI = \frac{CaO - MgO + BaO + SrO + K_2O + Na_2O + Li_2O + V_2 (MnO + FeO)}{SiO_2 + V_2 (Al_2O_3 + TiO_2 + ZrO_2)} \quad \dots(5)$$

Many researchers have reservations about using a basicity index (14-17) for predicting weld metal quality. The fact that the basicity index can be used to predict weld metal oxygen content as suggested in the literature is only a limited correlation for fluxes that are made up of primarily MgO, CaO, CaF₂ and SiO₂. For reliable prediction of weld metal oxygen content in weldments prepared with other complex flux systems that contain major additions of Al₂O₃, TiO₂, FeO and MnO new weld metal oxygen index needs to be developed.

It was reported that in general the higher the basicity, the fewer the nonmetallic inclusions in the weld metal. Eagar (18) found that for the fluxes investigated the weld metal oxygen content drops from about 900 to 250 ppm for a basicity index change from 0.5 to 1.5 and then remains constant at the 250 ppm level with further increases in basicity.

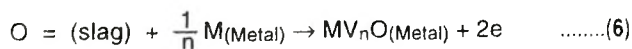
The tendency of a low-alloy steel to pick up oxygen from a slag can be shown using thermodynamic calculations to be directly related to the thermodynamic activity of FeO at the slag-metal interface. This activity is high for FeO-SiO₂, lower for MnO-SiO₂ and much lower for CaO-SiO₂ slags, though they may all have the same basicity. Consequently, basicity

index cannot represent the tendency of the metal to pick up oxygen from the slag.

The concept of basicity should be replaced by more fundamental concepts that will enable one to predict chemical activities of the alloying elements in solution (19). These in turn when combined with the thermodynamic free energies of formation of the components, should define the chemical potential and hence reactivity of any solution component. Recently optical basicity approaches have been explored as a means to predict weld metal oxygen (20). The optical basicity index reports the ratio of the electron donor power in an oxide system to the electron donor power of free oxide anions. This optical basicity index can be measured with a spectroscopic technique and has been found to correlate well for some flux systems to the weld metal oxygen content. Further improvement in carbon equivalent expression for weld metals should contain a more fundamental term such as the optical basicity index.

Influence of Welding Parameters

Electrochemical reactions have recently been identified to be as important as the thermochemical reactions in direct current welding (19,21-23). The electrochemical perturbation followed by a partially completed back thermochemistry in the weld pool, suggests a strong influence of the welding parameters on the weld metal composition and properties. Frost et al.(23) investigated the different chemical effects at anodes and cathodes. When direct current electrode positive (DCEP) welding with a silicate flux, the voltages are sufficiently high that all the slag constituents could decompose electrochemically. The electrolyses involved can be from the flux and/or the plasma. At the anode, the typical half cell reaction is :



The most stable oxides are preferred. The oxygen builds up in the liquid tip which detaches and falls into the weld pool. This process can cause a major perturbation in the weld pool oxygen content.

At the weld pool (cathode) the general half cell reaction is :



The reducible ions lost at the slag-weld pool interface can be replenished by diffusion through the diffusion boundary. The slag composition at the boundary layer will thus have a higher concentration of ions with large diffusion coefficients (i.e. basic constituents) than the bulk composition. Consequently, low valent basic oxides will predominate and high valent acid oxides will be present at lower concentrations at the interface in this diffusion-limited reduction. The net result will be a relatively large electrolysis of the more reducible oxides to form metals, which will then form alloys with the weld pool. Because of the chemically metastable alloy produced at the interface, there will also be a chemical back reaction that will limit this effect.

Kim et al. (21,24) demonstrated this electrochemical effect through chemical analysis of the electrode tip, detached droplets, and the weld metal as a function of travel speed of the weld pool. Electrochemical reduction can be more important in influencing metallic additions to the weld pool from the slag than thermochemical reactions. It is anticipated that influence of polarity and current needs to be better understood. This suggests that polarity, voltage, current and travel speed will have influence on weld metal oxygen which are not considered in current carbon equivalent or basicity index equations.

Influence of Thermal Experience

Most carbon equivalent expressions are only a function of composition and do not consider thermal experience and severity of quench. The thickness of plate, edge preparation and heat input of the process will influence the cooling rate. This can be schematically shown in the continuous cooling transformation (CCT) diagram in Figure 1. The cooling curve in Figure 1 is a function of the nature of the heat source as well as the heat extraction conditions. The time to cool from 800 to 500°C $\Delta t_{8/5}$ is an alternate way to describe the cooling rate of a weldment and has been used to compare the

welding thermal cycle to the acceptable thermal experience to achieve properties.

A formula devised by Lorenz and Duren (25) for calculating heat affected zone hardness, which takes into consideration both alloy composition and cooling rate, is shown in the following equation.

$$HV = 2019 \left[(1 - 0.5 \log \Delta t_{8/5}) C + 0.3 (CE - C) \right] + 66 (1 - 0.8 \log \Delta t_{8/5}) \dots (8)$$

and

$$CE = C + \frac{Si}{11} + \frac{Mn}{8} + \frac{Cr}{5} + \frac{Mo}{6} + \frac{V}{3} + \frac{Ni}{17} + \frac{Cu}{9} \dots (9)$$

Yurioka (26) also introduced a carbon equivalent type expression to describe low carbon steel weldability that has $\Delta t_{8/5}$ as a contributing term. The maximum heat affected zone hardness was related to the alloy composition and cooling rate.

$$H_{max} = 406C + 164CE_i + 183 - (369C - 149CE_i + 10) \arctan(x) \dots (10)$$

Where

$$x = \frac{(\log \Delta t_{8/5} - 2.822 CE_i \pi = 0.262)}{(0.526 - 0.195 CE_i \pi)} \dots (11)$$

and CE_i , CE_{II} are expression related to the chemical composition of the alloy that have identical format of the IIW carbon equivalent equation.

These are some of the expressions that stress both hardenability and weld thermal cycle in predicting weld metal mechanical behavior. An approximate calculation for $\Delta t_{8/5}$ can be obtained from the Rosenthal solution for welding heat behavior (27). $\Delta t_{8/5}$ can be directly related to both the heat input, H, and preheat temperature T_0 by the following equations (28) :

$$\Delta t_{8/5} = 8.149 \times 10^{-4} \left(\frac{\eta H}{2\pi\kappa} \right) \dots (12)$$

and

$$\Delta t_{8/5} = 2.767 \times 10^{-6} \left(\frac{\eta H^2}{4h^2\pi\kappa\rho C_p} \right) \dots (13)$$

In these equations, η and H are the efficiency and heat input of the welding process, κ , ρ , and C_p are

thermal conductivity, specific gravity and specific heat of the material, h is the thickness of the plate. Equation 12 is for three dimensional heat transfer conditions such as those found in thick plate welding while Equation 13 is intended for two dimensional heat flow in thin plate welding.

It is clear that to describe the microstructure and mechanical behavior of a high strength low alloy steel weld metal, the effects of alloying elements, oxygen, welding parameters, and cooling rate need to be considered. A generic weld metal carbon equivalent expression should therefore include the following terms.

$$CE_{max} = F(Mn, Si, \dots) + F(O_{inclusions}) = F(V, I, s, Polarity) + F(\Delta t_{8/5}) \dots (14)$$

$F(Mn, Si, \dots)$ is the term that describes the alloying elements hardenability effect. The complex interaction of weld metal inclusions with phase transformations is represented by $F(O_{inclusions})$. The $F(V, I, s, Polarity)$ term considers weld metal chemical composition alterations due to electrochemical reactions. Microstructural changes due to heat input and cooling rate change is expressed in $F(\Delta t_{8/5})$.

Fundamental Approaches

Liu et al. (29) have considered more fundamentally derived forms of the predictive equations. They evaluated both thermodynamic and kinetic approaches to the formulation. Short descriptions of the derived equations and their suggested use are given below :

Thermodynamic Approach

A metallic solution may be defined as a homogeneous phase composed of a mixture of different chemical elements, whose concentrations may be varied without the precipitation of a new phase. Austenite and ferrite are metallic solutions composed mainly of iron, with the addition of carbon and other alloying elements such as manganese and silicon. As metallic solutions, austenite and ferrite can be described by thermodynamic functions, the integral free energy of a solution can be written as :

$$G_{sol} = \sum_{i=1}^a X_i \mu_i^0 + RT \sum_{i=1}^n X_i \ln X_i + RT \sum_{i=1}^n X_i \ln \gamma_i \quad \dots\dots\dots (15)$$

where X_i are the mole fractions of the different components of the system, μ_i^0 are the standard partial molar free energies of the components, γ_i are the activity coefficients of the elements, R is the universal gas constant, and T is the temperature of the system.

In the case of the austenite (γ) to ferrite (α) transformation, the free energy change involved can be written as :

$$\Delta G_{\gamma-\alpha} = \sum_{i=1}^n \theta_i X_i^\alpha + \sum_{i=1}^n \psi_i X_i^\alpha \ln X_i^\alpha \quad \dots\dots\dots (16)$$

where θ and ψ are related to thermodynamic properties of the solutes, X_i^α and X_i^γ are related by the element partition coefficient, f_i as described in the following equation.

$$X_i^\gamma = f_i X_i^\alpha \quad \dots\dots\dots (17)$$

Thus, if these coefficients can be determined, it is theoretically possible to understand the fundamental role of each element in solution.

For athermal transformations, where phase stability can be expressed by $\Delta G_{\gamma-\alpha}$, a microstructural sensitive property such as CE can be given by the following:

$$CE = K_0 [C + K_{Mn}Mn + K_{Si}Si + \dots K_C'CLnC + K_{Mn}''MnLnMn + K_{Si}''SiLnSi +] \dots\dots\dots (18)$$

where K_0 is a proportionality constant and $?$ are coefficients for the various alloying additions and are subject to fundamental interpretations as indicated in Equations 16. Mn, Si, C, \dots are concentrations of the different elements expressed in weight percent. This expression is based on the assumption that microstructural sensitive properties are directly related to the amount of alloying elements present in the initial and transformed phases. Notice the presence of both linear and nonlinear terms in Equation 18. Omission of the nonlinear terms will simplify the equation to a form similar to the IIV carbon equivalent equation, Equation 1. In the case of low alloy steels, omission of the nonlinear terms may not be too harmful and the coefficients could

be manipulated to compensate for the omission. However, it becomes apparent that in high alloy systems, some of the information about alloying behavior may be lost without the nonlinear terms.

Kinetics Approach

For a transformation to occur, the criterion of phase stability alone may not always be sufficient for the prediction of alloy behavior. For example, in austenite decomposition by diffusional processes, carbon transport is one of the most important phenomena to be considered. Therefore, it may be expected that the carbon equivalent equations are related to the thermodynamic driving force for carbon transport. Let ΔG_p be the free energy change resulting from the difference in chemical potential for carbon in the two phases, $\Delta \mu_C$ i.e.,

$$\Delta G_p = N_C \Delta \mu_C \quad \dots\dots\dots (19)$$

where N_C is the number of moles of carbon. By definition, chemical potential of an element can be written as,

$$\Delta \mu_i = \frac{\delta}{\delta n_i} (\Delta G)_{T, P, n_i} \quad \dots\dots\dots (20)$$

and if the difference in free energy between the two solutions is known, the chemical potential difference for carbon, and consequently, the driving force of carbon transport can be determined.

By considering whether or not there is partitioning of alloying elements during austenite decomposition, two groups of solutions can be developed. In the first group, $X_i^\alpha = X_i^\gamma$ and the cooling rate is assumed to be high enough to impede any atomic diffusion process from occurring. For some of the alloying elements, with larger size atoms and more sluggish atomic transport such as Cr and Mo, this may be an appropriate assumption. With partition coefficients equal to one, free energy change of austenite to ferrite transformation can be written as:

$$\Delta G_{\gamma-\alpha} = \sum_{i=1}^a X_i^\alpha (\mu_i^\alpha - \mu_i^\gamma) + RT \sum_{i=1}^n X_i^\alpha \ln \frac{\gamma_i^\alpha}{\gamma_i^\alpha} \quad \dots\dots\dots (21)$$

Carrying out the differentiation and substituting the result in Equation 19, the following expression is obtained after simplifying:

$$\Delta G_p^c = X_c [\lambda c + \sum_{i=1}^a \mu_i - X_i] \dots \dots \dots (22)$$

Assuming that the carbon equivalent can be directly related to the thermodynamic driving force for carbon transport, ΔG_p^c , a new form of the equation for carbon equivalent can be obtained. In Equation 22, X_i and X_c are expressed in mole fractions. By introducing common alloying elements, the hardenability formula, or CE expression, can be expressed as:

$$CE = k_0^c [1 + k_c^c c + K_{Mn}^c Mn + K_{Si}^c Si + \dots] \dots (23)$$

The coefficients K_i^c also contain conversion factors for the mole fraction to weight percent change. Notice that Equation 23 is different from Equations 1 to 4 and the form suggests by the multiplication relationship the interaction between carbon and other alloying elements. The presence of interaction terms is reasonable since each alloy addition influences the carbon behavior and thus should be a product term with carbon. This new suggested form of the carbon equivalent equation should better fit the low carbon microalloyed steels where there is carbo-nitride precipitation.

In some transformations, however, the consideration of $X_i^a = X_i^g$ may not be proper. It is known that even in para-equilibrium conditions, carbon is observed to partition. To illustrate the difference due to the situation of $X_i^a = X_i^g$, the $\Delta G_{\gamma-\alpha}$ equation (Equation 16) is reconsidered without assuming the non-partitioning behavior. Following the same procedure illustrated previously, the following expression is developed.

$$\Delta G_p^c = X_c [P_c + \sum_{i=1}^a P_i X_i + \psi L_n X_c + \sum_{i=1}^a X_i L_n X_i] \dots \dots \dots (24)$$

In terms of Equation 22, the carbon equivalent can now be given as

$$CE = K_0^c [1 + K_c^c c + K_{Mn}^c Mn + K_{Si}^c Si + K_{LC}^c Ln c + K_{LC}^c CL_n c + K_{Mn}^c Mn Ln Mn + K_{Si}^c Si Ln Si + \dots] \dots \dots (25)$$

Notice that the equation is again different from the commonly used form of carbon equivalent expressions as seen in Equations 1 and 2. In addition to the terms expressing the interaction between elements as suggested in Equations 18 and 23, terms containing the natural logarithm of element concentration are also present. The last three terms are a major concern when there are orders of magnitude variations for minor alloying additions. The lack of these terms may cause problems in the efforts to correlate microalloyed HSLA steel properties to composition. In addition to the elements considered above, the oxygen content must be included to make these expressions useful in predicting weld metal properties.

Isoproperty Diagram

As shown previously in Equations 8 to 11, investigators have modified the carbon equivalent equation to include the influence of the cooling rate, $T_{8/5}$. This is the realization that aside from alloy composition, the selection of welding process parameters can also alter the weld microstructure and properties. A diagram, which correlates the thermal experience and hardenability to weld metal microstructure, would be very useful in the selection of welding consumables for different welding conditions, heat input, and alloy composition. Horii et al.(30) recently suggested a $D_{8/5}$ versus carbon equivalent diagram to predict weld properties, as shown in Figure 2. The line represents welds with equal toughness that resulted from different welding conditions and alloy composition. This figure shows that even with process modifications or new materials selection, the desirable microstructure can be predicted using the comprehensive carbon equivalent equations suggested by Duren and Yurioka (25-26). The concept of using these types of isoproperty diagrams which represent the total welding experience, both thermal and chemical, would be a major improvement in our ability to design a weld metal and quantitatively predict its properties by specifying the correct welding parameters and consumables. The suggested mathematical forms should be considered for the carbon

equivalent expression to be used with the isoproperty diagrams.

Summary

Carbon equivalent type expressions, traditionally used in describing microstructure and properties of wrought steel products, are generally inadequate in describing weld metal phase transformations and cracking susceptibility. Oxygen and inclusions affect significantly the kinetics of austenite-to-ferrite transformation in the weld metal. In addition to the thermochemical reaction that occur in the weld pool, the once considered unimportant electrochemical reactions also adjust the final weld metal chemical composition. New forms for the carbon equivalent expressions should be applicable to a wide range of welding parameters and alloy composition. Fundamental thermodynamic and kinetic approaches were used to obtain predictive equations with better ability in predicting weld metal behavior. Isoproperty diagrams correlating welding process parameters and alloy composition showed excellent promise in weld metal properties prediction and selection of welding processes, parameters and consumables.

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