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Inclusion formation in low-alloy steel welds†

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Introduction

In recent years, modeling reactions in liquid and solid state during welding have provided greater insight to microstructure development in welds, that could not be obtained otherwise.[1,2] These models must consider all physical processes and describe their interactions as a function of thermal cycles at different regions and the composition. The present work pertains to formation of inclusions in the weld metal region.

Inclusions form in steel welds as a result of deoxidation reaction between dissolved aluminum, titanium, silicon, and manganese with dissolved oxygen. The inclusion characteristics such as volume fraction, size, number density, composition and the type of surface compounds have influence on the subsequent solid-state transformations.[2-4] For example, Ti-rich inclusion are expected to promote the formation of acicular ferrite (Fig. 1). Predominant acicular ferrite transformation from austenite was initiated by the addition of 28 wt. ppm Ti to a Fe-C-Si-Mn weld. In the absence of Ti, only bainitic microstructure was observed.[3]

This relationship between acicular ferrite and inclusion characteristics have been found in many other cases.[3,4] However, the predictive methodology to induce the proper inclusion type for a given application and produce a specific weld microstructure, does not exist. Therefore, we need to understand the inclusion formation in steel welds as a function of weld metal composition, welding processes, and process parameters.

In this paper, we review (1) models to describe the inclusion formation, (2) experimental work to understand the inclusion formation in rapid weld cooling conditions and evaluation of an inclusion model; and (3) future research on inclusion formation.

Models for Inclusion Formation

In the past ten years much effort has been focused on developing models for inclusion formation. All these models start with an overall weld metal composition and describe the inclusion characteristics such as volume fraction, size, number density, composition, and oxidation sequence.

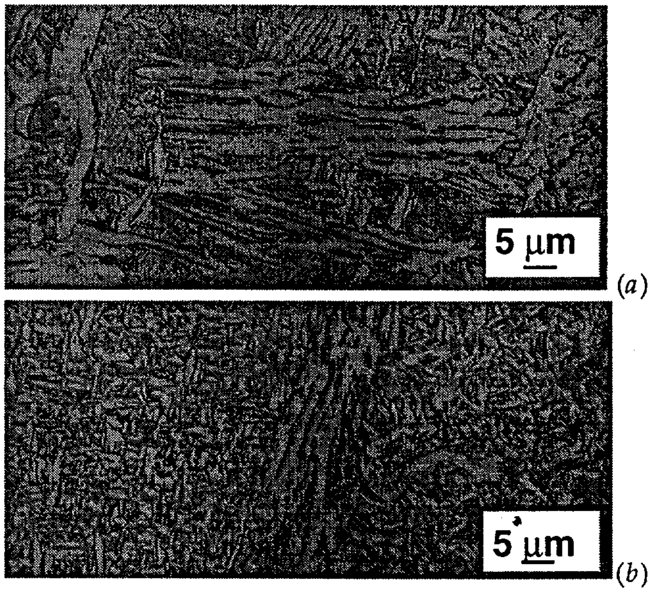

Fig. 1. Presence of titanium promotes the acicular ferrite reaction in steel weld metals: (*a*) 7 wt. ppm Ti and (*b*) 28 wt. ppm of Ti.[3]

Fixed Oxidation Sequence Method: Early work by Kluken and Grong was based on the nominal weld composition, mass balance and fixed oxidation sequence.[5] This work was later adopted by Bhadeshia *et al.* in their model for weld microstructure.[2] In this methodology, the volume fraction of inclusions was estimated using oxygen and sulfur concentrations.

The inclusion formation sequence was fixed as follows: (1) dissolved aluminum in the liquid steel reacts to form Al_2O_3 and aluminum nitride (AlN) formation is considered to be improbable; (2) Titanium reacts with nitrogen to form TiN; (3) Residual titanium after the formation of TiN reacts with oxygen to form Ti_xO_y type of oxides; (4) Residual oxygen reacts with manganese and silicon to form MnO and SiO_2. With the above sequence the inclusion composition was estimated. However, this methodology requires experimental measurements of residual Al and Ti in matrix to estimate the extent of Al_2O_3 and Ti_xO_y reactions.

The above method is simple to use and gives a description of inclusion composition as a function of nominal composition and has been shown to be in good agreement with the experimental measurements.[5] However, the limitations of this methodology are grouped into two categories as following.

Weld metal composition effects: The fixed oxidation sequence is not valid for a wide range of weld metal compositions. This methodology ignores the variation in sequence with a change in concentration of Al, Ti, Si, Mn, and O. Therefore, some of the experimental observations of inclusion morphology can not be explained. Moreover, the formation of complex oxides is not considered and experimental measurements of residual Al and Ti in solid solution are needed.

Process effects: The process condition has a strong influence on the inclusion formation and is not considered in this methodology. As a result, the inclusion number density and size can not be estimated. Moreover, the effect of heat transfer and fluid flow on the inclusions is not considered.

The inclusion modeling efforts subsequent to the above work have tried to address either one or both issues mentioned above. The weld metal composition effects have been addressed with multicomponent, multiphase thermodynamic calculations. The process effects have been addressed by coupling thermodynamic and kinetic calculations.

Thermodynamic Model: Multicomponent, multiphase thermodynamic calculations between liquid steel, simple oxides, and complex oxides to describe inclusion formation can be performed by thermodynamic softwares.[6] Hsieh *et al.*, described the inclusion formation with Scheil-type calculations in the temperature range of 2300 to 1800 K.[7] and showed that both simple and complex oxides form at different temperatures. The sequence of oxides, type of oxides and formation temperature were found to vary with weld metal composition. These calculations also allowed to estimate the residual aluminum and titanium in solution. This work essentially showed that the inclusion formation is sensitive to weld metal composition and temperature.

In a recent work, Koseki *et al.* carried out multicomponent, multiphase thermodynamic calculations extending beyond the solidification and up to solid state regions.[8] Their calculations indicated that there may be complex phase change in-between oxides that form in the liquid steel. For example, their calculations in a Fe-C-Si-Mn-Al-Ti steel weld showed that at high temperature (> 1600 K) Ti_3O_5 and $3Al_2O_3.2SiO_2$ may form from liquid steel and at low temperature (<1600 K) $3Al_2O_3.2SiO_2$ may decompose to form $MnO.Al_2O_3$. Further, for this reaction to occur there must be depletion of Mn from the austenite surrounding the matrix. An additional calculation showed that fraction of glassy oxide is reduced due to addition of titanium. Therefore, Koskei *et al.* concluded that this might be the reason for acicular ferrite nucleation on the inclusions.

Similar calculations are being performed by Van der Eijk et al. to investigate the effect of oxides on solid state transformation in conventional steels.[9] Recently, Blais et. al. used thermodynamic calculations based on oxide solutions instead of simple stoichiometric oxides.[10] These calculations suggest the presence of liquid inclusions within the austenite. The mechanism for the above type of inclusion formation remains uncertain. However, thermodynamic calculations do not consider the effect of process conditions on the kinetics of these reactions at various temperatures.

Ichikawa et. al.[11] extended the thermodynamic calculations by Koseki et. al. to estimate the number density of inclusions.[8] This was done by assuming that the inclusions have an average diameter of ~0.5 μm and do not vary with weld metal composition or process. However, this assumption may not be valid for wide range of weld heat input conditions and processes.

Thermodynamic methods described above are capable of illustrating the complex effects of weld metal compositions and temperature. They can be used for evaluating the chemical effects and may provide useful information for weld consumable development. However, they are not capable of describing the effects of process conditions.

Thermodynamic-Kinetic Model: During weld cooling, many phases (oxides, sulfides, and nitrides) can form simultaneously and the kinetics of one reaction can interfere with the kinetics of the other reactions. There are no established methods to consider this simultaneous reaction kinetics. Recent work has shown that the simultaneous reaction kinetics can be handled with modified Johnson-Mehl-Avrami type equations.[12] However, parameters that are needed for this approach need experimental data which are not available for reaction kinetics in the liquid steel.

However, with some assumptions overall oxidation kinetic equations can be extended to describe inclusion formation in steel welds. Babu et. al[13,14] have developed an inclusion model with some assumptions. The details are given below: (1) In the model, only the formation of simple oxides were considered. (2) At a particular temperature, thermodynamic equilibrium between liquid steel and only one stoichiometric oxide is considered, ignoring the effects of other phases; (3) The calculations consider the effect of all alloying elements (C, Si, Mn, Ni, Mo, Cr, V, Co, Ti, Al, Cu, W, Zr, O, N, and S) to calculate the activities of each element and is consistent with multicomponent thermodynamic calculations between two phases; (4) The oxide to form first is determined by the magnitude of its driving force for formation, ΔG, and is assumed to nucleate homogeneously. Subsequent oxides are assumed to form heterogeneously on the first-oxide; (5) To model the reactions during continuous cooling, additivity of volume fractions is assumed. By this assumption, the volume fraction of inclusion forming at different temperatures are added together. The kinetic calculations are modified accordingly. The weld cooling curves were modeled using the equation given by Ion et al.[15]

The inclusion model developed with the above assumptions has been shown to be very sensitive to weld metal composition and weld cooling conditions.[16] Evaluations of this model for a wide range of welding processes and weld cooling conditions are currently being pursued.[16,17]

This thermodynamic-kinetic inclusion model, however, has the following limitations. Since the model does not consider multi-phase equilibria and complex oxides, the model does not predict the formation of Mn-rich oxides such as $3Al_2O_3.2SiO_2$, $MnO.SiO_2$ and $MnO.TiO_2$. Preliminary calculations, show that complex $MnO.Al_2O_3$ do form and can be considered.[17] The inability to consider Mn-rich inclusion leads to under prediction of Mn concentrations in the inclusions. The assumption of stoichiometric oxide formation needs further attention. Moreover, this inclusion model does not consider effects of solidification partitioning and fluid flow on inclusion growth.

Thermodynamic-Kinetic-Fluid Flow Model: A recent experimental investigation in steel melts has shown that collision and coalescence of inclusions may lead to rapid growth of inclusions (Fig. 2).[18] The collision and coalescence of inclusions can be promoted by fluid flow velocity gradients in the weld metal region. In addition, the fluid flow conditions in the weld may transport inclusion to regions with varying temperature. This may lead to complex dissolution and growth of inclusions. Hong *et al.* have used computational heat transfer and fluid flow models to describe this type of inclusion growth.[19] In this model, the growth and dissolution of only Al_2O_3 oxide was considered. The calculations show 66% of inclusion population will experience complex temperature oscillations and, therefore, will undergo dissolution and growth. In addition, preliminary calculations have been performed to consider the fluid flow velocity gradients. The above approach has the potential for simulating the realistic conditions of inclusion formation in steel welds. However, further work is necessary to consider the complex oxide growth, solidification effects and partitioning of the inclusions to the slag layers.

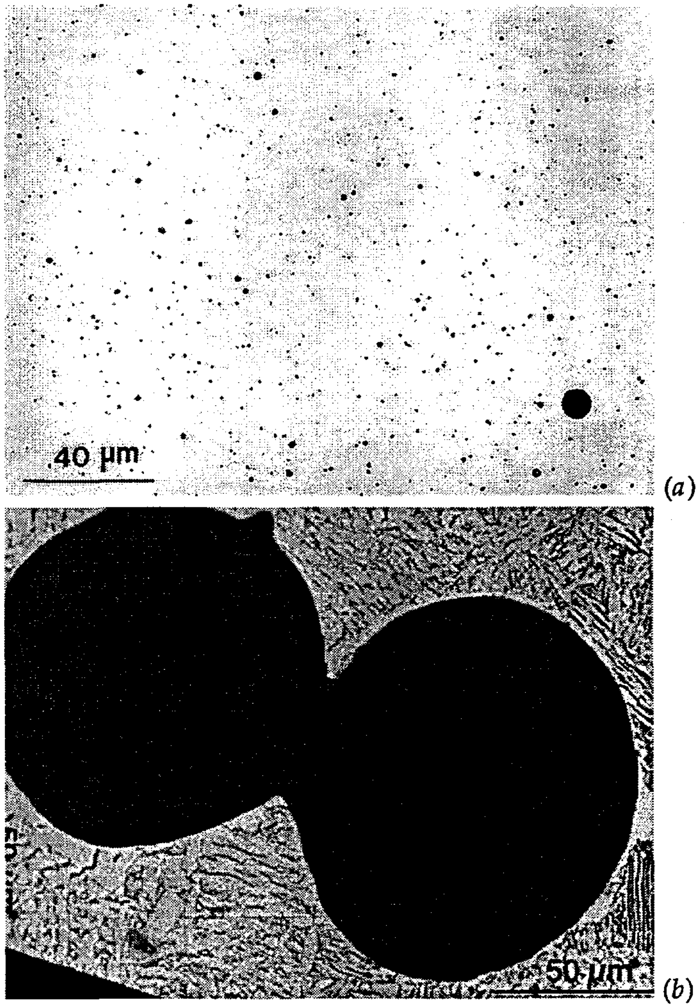

Fig. 2. Effect of fluid flow velocity gradients on the inclusion growth: (*a*) as welded condition; (*b*) after holding at 1480 °C for 60 s with fluid flow velocity gradients in a Gleeble thermo-mechanical simulator.[18]

Application to Welding Processes

Most of the models discussed above were applied to practical weld consumable development work. Examples of this work are discussed briefly.

Thermodynamic Calculations: Inclusion formation and microstructure evolution in self-shielded flux cored arc welds are being studied.[20] These welds were produced with flux cored wires having large aluminum contents to promote deoxidation. Final weld metal composition of the weld is Fe - 0.234 C - 0.50 Mn - 0.28 Si - 1.77 Al - 0.003 Ti - 0.006 O - 0.064 N (wt.%). Some of the welds were produced comparatively at low levels of aluminum in the filler wire and typical weld metal composition was Fe - 0.149 C - 0.64 Mn - 0.30 Si - 0.53 Al - 0.058 Ti - 0.030 O - 0.033 N (wt.%).

The weld microstructure in high aluminum and low aluminum welds were found to be different. In the case of high aluminum welds, the weld metal microstructure showed skeletal ferrite network due to incomplete transformation to austenite from δ ferrite on weld cooling. In contrast, low aluminum welds showed classical allotriomorphic and acicular ferrite microstructure. Optical and electron microscopy showed that there is a competition between Al_2O_3 and AlN formation in these two welds and this was supported by thermodynamic calculations.

Further work is necessary to describe this heterogeneous nature, the inclusion number density and size of these inclusions and also the slag-metal reactions in these welds.[21] The above example illustrated that thermodynamic calculations can indeed describe gross changes in weld inclusion formation.

Thermodynamic-Kinetic calculations: The inclusion formation and microstructure development in submerged arc (SAW), electron beam (EB) and laser beam (LB) welds were investigated.[16] The nominal composition of the SAW welds was Fe - 0.1 C - 1.61 Mn - 0.81 Si - 0.015 Al - 0.018 Ti - 0.084 O (wt.%) and the heat input was 1.87×10^6 J m^{-1}. Autogenous electron beam (heat input = 0.653×10^6 J m^{-1}) and laser beam (heat input = 0.184×10^6 J m^{-1}) welds were made on top of the submerged arc welds. For EB and LB processes, with a decrease in the heat input, cooling rate in the weld metal region is expected to increase. The increased cooling rate may lead to changes in the inclusion characteristics.

Typical inclusion distribution is shown in Fig. 4. Experimental measurements showed the following results: (1) Oxygen loss in EB (0.037 wt.% O) and LB (0.053 wt.% O) welds from the original oxygen level (0.084 wt.% O) in SAW weld was observed; (2) Inclusion volume fraction and diameter decreased (see Fig. 4); (3) The measurements from high-magnification indicated that inclusion number density increased; and (4) Inclusions from EB and LB welds were rich in Al and Ti.

The oxygen loss from the EB and LB welds were expected due to low partial pressure of oxygen in the atmosphere above the weld pool. The oxygen loss is expected to reduce the inclusion volume fraction and diameter and number density. However, the number density of inclusion increased in EB and LB welds. It was speculated that this result is due to competition between the effects of reduced driving force for oxide formation (due to oxygen loss) and the increased undercooling for inclusion nucleation and growth (due to rapid weld cooling in EB and LB welds). The above hypothesis was evaluated with the thermodynamic-kinetic inclusion model.[13,14]

Approximate weld cooling curves were calculated using the method given by Ion *et al.*[15] In this calculation, the effect of weld pool shapes were ignored. The calculated inclusion volume fraction, diameter, and number density are compared with measured data (Fig. 5). The inclusion model predicted the drop in volume fraction and diameter with EB and LB welds. Moreover, the inclusion model predicted the increase in number density of inclusions. Although calculated trends agree with the experimental observations, the magnitudes of inclusion number density and diameter do not agree with the measured values. This is related to the approximate weld cooling rates used in the calculations.

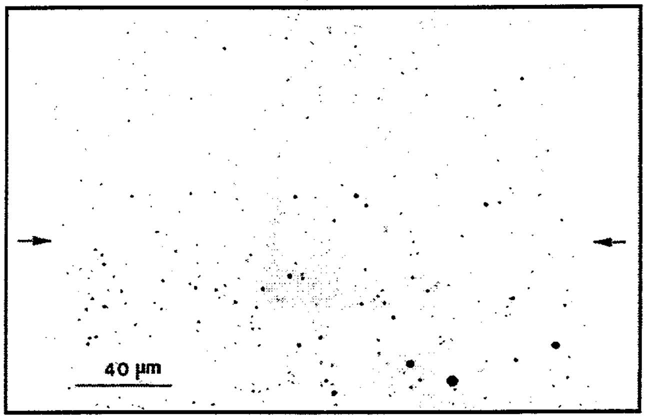

Fig. 4. Unetched optical micrograph illustrating the transition region between SAW weld (bottom) and EB weld (top) regions. A change in inclusion size distribution is evident.

Nevertheless, the calculations show crucial information to understand the inclusion formation in EB and LB welds. The calculations show that the first oxide was Al_2O_3 in all three welds. However, the temperature of completion for Al_2O_3 (or in other words Al_2O_3 formed at large undercooling) was found to be lower in the case of EB (2075 K) and LB (2095 K) welds compared to SAW (2129 K) welds. This resulted in the increased number density values for EB and LB welds.

The calculations also showed that in EB and LB welds, the inclusions would be made up of mostly high temperature oxides and predicted the inclusions will be enriched in Al and Ti. This is in agreement with the experimental observations. However, the silicon and manganese concentrations in the inclusions did not match with the experimental observations. This is attributed to the formation of complex $MnO.SiO_2$ and $MnO.TiO_2$ reactions in the liquid steel and also during solidification. The above example showed that thermodynamic-kinetic model has general applicability. Further evaluations in high strength steel welds also showed similar results.[17]

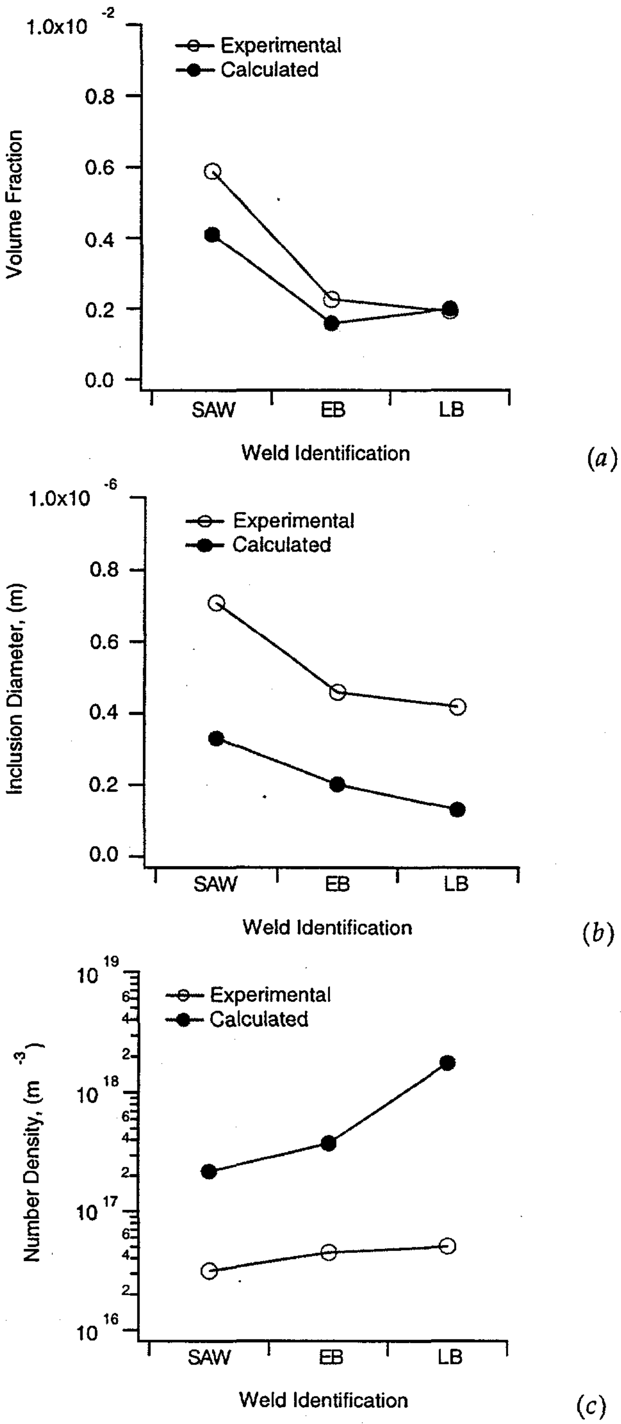

Fig. 5. Comparison of calculated and experimental inclusion characteristics in SAW, EB and LB welds. (*a*) inclusion volume fraction; (*b*) inclusion diameter and (*c*) inclusion number density.

Future Work

Further improvements in the inclusion modeling efforts must address various issues as following: (1) multicomponent, multiphase thermodynamic equilibria with kinetic considerations need to be incorporated. Specifically, calculations need to consider solutions of oxide and the mechanisms of the formation and trapping within the solid.[10] (2) Inclusion model must consider the reactions during solidification considering the solidification rates and related partitioning effects. The model needs to be extended to consider the feasibility of reactions in the solid state. (3) Inclusion models must be coupled with computational heat transfer and fluid flow models so that they can describe the effects of heat transfer and fluid flow rigorously. For example, the effect of inclusion growth and dissolution due to temperature fluctuations need to be (see Fig. 6) considered.[19] The partitioning of these inclusions to slag need to be incorporated. (4) Effect of these inclusions on acicular ferrite formation need to be investigated and operating mechanisms need to be evaluated for various types of inclusions. The mechanisms of local depletion of carbon or manganese around the inclusions need to be evaluated with kinetic considerations such as diffusion, weld cooling rates and thermal cycling.

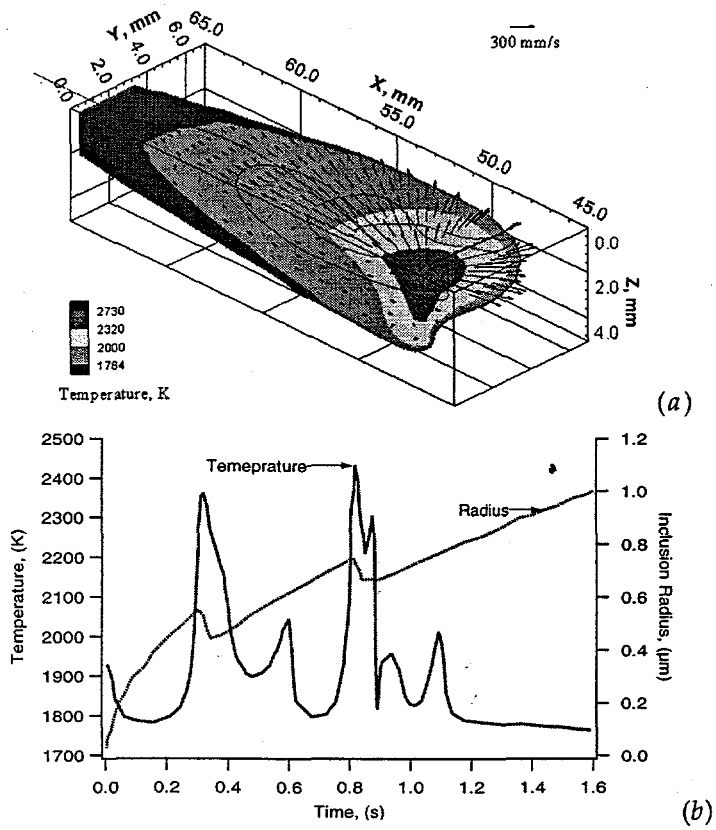

Fig. 6 (*a*) Temperature and velocity fields and typical path of particle in weld pool. (*b*) Typical Al_2O_3 inclusion temperature fluctuations and corresponding size change in weld pool.

Summary and Conclusions

This paper summarizes four models for describing inclusion formation in steel welds. These methods include simple fixed oxidation sequence model, thermodynamic model, thermodynamic - kinetic model, and thermodynamic - kinetic - fluid flow model. Complexities of these models increase with a need to describe details of the inclusion formation. The applicability of the models was illustrated with two examples.

In one of the examples, thermodynamic calculations of phase stability between liquid steel, AlN and Al_2O_3 explained the inclusion formation in self-shielded flux cored arc weld. In the second example, thermodynamic-kinetic calculations illustrated the competing effects of weld metal composition and cooling rate effects on the inclusion formation in electron beam and laser beam welds.

Limitations of the current inclusion models and recommendations for future work on the inclusion formation were highlighted.

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