



Study the Thermodynamic Behaviour of Inclusions in High Heat Input Welding Steel

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Abstract

Investigation on the thermodynamic precipitation behavior of inclusions is a prerequisite for achieving the fine and dispersed distribution of inclusions in steel. To clarify the influence of cooling rate on the formation pathway, precipitation pathway evolution and precipitation conditions of complex inclusions in EH40 ship plate steel, this paper systematically investigates the precipitation behavior of inclusions by combining FactSage with the Won-Thomas microsegregation model. The results show that Mg is the key element promoting the transformation of Al_2O_3 to spinel-type composite oxides; it is difficult to form stable Al-Ti composite oxides directly between Al_2O_3 and Ti_2O_3 , and Ti mainly enters the composite inclusion system with the participation of Mg. Al, Mg and Ti all segregate and enrich in the interdendritic residual liquid phase during solidification, and local supersaturation promotes oxide precipitation. Al_2O_3 satisfies the precipitation condition at the early stage of solidification, while Ti_3O_5 and Ti_2O_3 reach their precipitation conditions at similar stages. This study provides important theoretical guidance for regulating the modification process of inclusions in steel.

Keywords: Ship plate steel, Complex inclusions, Cooling Rate, Oxide metallurgy

1. Introduction

High heat input welding steel is widely used in shipbuilding, petroleum, and pipeline steel, etc. However, the heat-affected zone of steel after high-energy welding with large line energy is prone to coarse grains and brittle fractures, causing hazards [1,2]. The oxide metallurgy technology regulates the fine and dispersed distribution of inclusions in steel, inducing the nucleation of needle-shaped ferrite within the crystal, which can effectively refine the grain structure and enhance the welding performance of the material [3].

In recent years, a large number of scholars have conducted research on the oxide metallurgy process and inclusions nucleation. In terms of production processes, Luo et al. explored the influence

of different elements and microalloying components (e.g., C/Ti/Mn) on the steel structure [4-7]. Wang et al. [8] studied the influence of different addition sequences of deoxidizers on the steel properties, proving that the composite deoxidation method of Mg-Al-Ti has the best organization refinement degree and the best low-temperature impact toughness. Cui X K et al. [9] studied the number density, size, distribution, composition and evolution laws of inclusions in steel under different S content and Al content. Under the condition of Al less than 0.0050%, the main oxide inclusions in the steel are $\text{MgO} \cdot \text{Al}_2\text{O}_3$, La_2O_3 and oxides of Ti. When the S content is around 0.0075%, inclusions have MnS precipitation on the outside, and the average size and number density of inclusions increase. When MnS precipitates on inclusions, the Al content is increased to affect the number density



and distribution of inclusions, thereby affecting the formation of IAF in the steel.

In terms of inclusions growth paths, Xiao et al. [10] and Li et al. [11] conducted in-depth studies on the stable structures and thermodynamic stability of magnesium oxides, aluminum oxides, titanium oxides and zirconium oxides clusters. Meng et al. [12-14] explained the adsorption structures and electronic properties of different composite inclusions. While most previous research efforts have focused on individual Mg-Ti/Mg-Al inclusions, relatively little research has been conducted on the microsegregation of composite inclusions. Some research into the deoxidation of Mg-Al-Ti composites also focuses on the sequence of deoxidiser addition and the production process.

In summary, the existing research mainly focuses on production processes and inclusions nucleation mechanisms. The cooling mechanism, as an important factor affecting material properties, has received little research on the regulation of inclusions precipitation behavior based on thermodynamics and kinetics. Hao X D et al. [15] studied the phase transformation, precipitation and mechanical properties of titanium microalloy steel under different cooling processes, and found that nano-carbonitrides were precipitated during cooling and played a precipitation strengthening role. Wei Jianfeng et al. [16] obtained Ohnaka's equation through a large number of microscopic segregation model studies and calculations, verifying its applicability to GCr₁₅ bearing steel and low-carbon steel. However, in high heat input welding steel, the Won-Thomas microsegregation model considers the limited solid-phase diffusion effect, which can more reasonably describe the solute redistribution behavior during the solidification process of actual steel liquid [17]. Its physical assumption lies between the fully balanced lever law and the highly non-equilibrium Scheil model, and more closely resembles the actual solidification conditions of steel. Unlike the Scheil and lever rule models, the Won-Thomas model is specifically designed for multi-component, multi-phase steel types and can predict solute redistribution and phase fraction evolution in ship plate steel more accurately during eutectic phase transformation. Additionally, this model can be calibrated using process parameters such as secondary dendrite arm spacing to obtain highly precise predictions of the solidification conditions in this study.

Based on this, this paper takes industrial production of EH40 ship plate steel as the research object. Firstly, FactSage is used to calculate the stable zone and the trend of complexification of the Al-Mg-Ti-O oxide system, clarifying the formation conditions of MgAl₂O₄ and Ti-containing composite oxides; Secondly, the Won-Thomas microsegregation model is used to analyze the enrichment laws of Al, Mg and Ti in the residual liquid phase between dendrites during solidification and their influence on inclusions precipitation; Finally, the solidification heat simulation

experiments and SEM-EDS characterization are carried out to verify the simulation results. This paper aims to clarify the influence mechanism of cooling rate on the formation path, complexification degree and size control of Al-Mg-Ti composite inclusions, providing important theoretical basis for the optimization of oxide metallurgy process and controlled design of inclusions in EH40 ship plate steel.

2. Description of the approach, work methodology, materials for research, assumptions, experiments etc.

2.1. Experimental materials

The experimental material is EH40 high-strength ship plate steel, which is produced industrially. The main production processes are converter smelting, LF furnace refining, magnesium treatment, titanium treatment and continuous casting. This steel is strong, tough and easy to weld, and is often used in ship and marine engineering structures. The main chemical composition of the experimental steel is shown in Table 1.

Solidification thermal simulation experiments were carried out at different cooling rates using a thermal simulation test machine for the continuous casting slab solidification process. Based on the principle of heat transfer similarity, the device can simulate the solidification, dendrite growth, and inclusion precipitation conditions of molten steel during the continuous casting process. The experimental steel was machined into a 100 mm × 5 mm × 4 mm sample using wire electrical discharge machining, then placed in a corundum crucible to minimise external contamination. During the experiment, the temperature was increased to 1853 K at a rate of 10 K/min and held for 60 minutes to ensure a uniform temperature throughout the sample and to eliminate the influence of the previous thermal history. The sample was then cooled to 1273 K at cooling rates of 100, 50 and 10 K/min, respectively, to correspond to fast, intermediate and slow cooling conditions. Each cooling condition was repeated at least three times.

After the thermal simulation experiment, the samples were cut and sampled along the cross-section. Following inlaying, grinding, polishing and cleaning with anhydrous ethanol, the morphology of the inclusions was observed using a scanning electron microscope and the distribution characteristics of O, Mg, Al, Ti and other elements within the inclusions were analysed using EDS.

Table 1.
Composition of the experimental steel (wt.%).

Elements	C	Mg	Al	Ti	Mn	S	P
Mass fraction (wt.%)	0.08	0.003	0.033	0.017	1.45	0.0025	0.008

2.2. Thermodynamics and microsegregation calculation method

Phase diagrams for the binary systems MgO-Al₂O₃, MgO-TiO₃ and Al₂O₃-TiO₃, as well as the multicomponent system Al-Mg-Ti-O, were calculated using FactSage 8.2 thermodynamic calculation software [18]. The stable regions and composite trends of the different oxides were analysed. The calculated temperature range was set to 1300–1900 K to cover the solidification temperature range of molten steel [19, 20]. By comparing the distribution of the different oxide phases, the thermodynamic possibility of forming Al-Mg-Ti-O composite inclusions was determined, as was the effect of Mg and Ti on the evolution of the inclusion composite.

The Won-Thomas microsegregation model describes the redistribution behaviour of solute elements, such as Al, Mg and Ti, between the solid and liquid phases during solidification. The Won-Thomas model is expressed as follows:

$$C_z = C_0(1 - (1 - 2\epsilon k)f_s)^{\frac{k-1}{1-2\epsilon k}} \quad (1)$$

In the formula, ϵ is the modified solid diffusion parameter, k is the distribution coefficient, and f_s is the solid fraction. The values of ϵ and k were adopted from Refs. [21] and [22], respectively, as they correspond to analogous solute-sorbent systems under comparable operating conditions. The solid fraction f_s was calculated based on the initial slurry composition.

According to the composition of the experimental steel, the liquidus temperature T_L and solidus temperature T_S were calculated by empirical formula :

$$T_S = T_0 - 344\omega(C) - 183.5\omega(S) - 124.5\omega(P) - 6.8\omega(Mn) - 12.3\omega(Si) - 4.1\omega(Al) - 18\omega(Ti) \quad (2)$$

$$T_L = T_0 - 90\omega(C) - 31.5\omega(S) - 32\omega(P) - 5\omega(Mn) - 6.2\omega(Si) - 3.6\omega(Al) - 18\omega(Ti) \quad (3)$$

In the formula, $T_0 = 1811$ K is the melting point of pure iron. Calculated $T_L = 1797$ K, $T_S = 1759$ K.

The main parameters required for the model calculation include the initial mass fraction of the element C_0 , the distribution coefficient k , the solid phase diffusion coefficient DS , the local solidification time t_f , and the secondary dendrite arm spacing λ . The relationship between the secondary dendrite arm spacing and the cooling rate is based on the empirical formula :

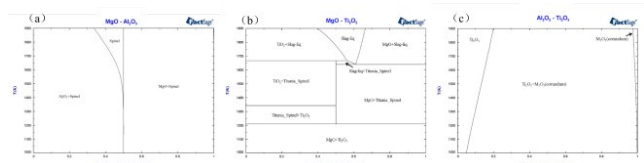
$$\lambda = (169.1 - 720.9\omega(C))\tau^{-0.4935} \quad (4)$$

In the formula, λ is the secondary dendrite arm spacing, μm ; τ is the cooling rate, K/min. The element activity was calculated by Wagner first-order interaction model. The local liquid phase composition after segregation was substituted into the oxide solubility product criterion to analyze the precipitation conditions of Mg, Al, Ti and other oxides under different solid fraction.

3. Description of achieved results of own researches

3.1. Thermodynamics and microsegregation calculation

In order to clarify the formation conditions of Al-Mg-Ti composite inclusions in the solidification process of EH40 ship plate steel, the thermodynamic calculation of the related oxide system was carried out by FactSage 8.2, and the results are shown in Fig.1.



(a) MgO-Al₂O₃ ; (b) MgO-Ti₂O₃ ; (c) Al₂O₃-Ti₂O₃

Fig. 1. Binary phase diagrams of oxide systems

Figure 1(a) shows the binary phase diagram for the MgO-Al₂O₃ system. The calculation results show that the MgAl₂O₄ spinel is stable over a wide range of conditions in the system. When the MgO/(MgO+Al₂O₃) molar ratio is between 0.35 and 0.50 and the temperature is between 1400 and 1900 K, MgAl₂O₄ spinel can exist stably. When Mg and Al coexist in molten steel, Mg can react with formed Al₂O₃ to create MgAl₂O₄ spinel inclusions. Compared with traditional Al₂O₃, MgAl₂O₄ has a more stable crystal structure and lower interface energy, which improves the morphology of inclusions and reduces aggregation. Therefore, in the Al-Mg composite deoxidation system, the transformation of Al₂O₃ to MgAl₂O₄ has an obvious thermodynamic driving force, and its reaction formula and standard Gibbs free energy are:

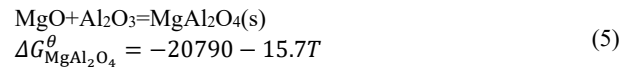


Figure 1(b) shows the binary phase diagram for the MgO-TiO₂ system. A narrow composite oxide phase region exists within the middle composition range of the phase region distribution of the system, encompassing both MgTiO₃ and MgTi₂O₄. The stable region of MgTi₂O₄ spinel is mainly concentrated within the MgO/(MgO+Ti₂O₃) range of about 0.40–0.65 and the temperature range of about 1210–1665 K. Its stable composition and temperature ranges are significantly narrower than those of MgAl₂O₄, and it is more sensitive to the Mg/Ti ratio and temperature changes. Figure 1(c) shows the Al₂O₃-Ti₂O₃ binary phase diagram. Within the solidification temperature range of molten steel, there is almost no solid-phase reaction between Al₂O₃ and Ti₂O₃; they exist as independent oxides. This demonstrates that, in an Al-Mg-Ti deoxidisation composite system, it is difficult to form a stable composite oxide solely from Al and Ti in the absence of Mg.

Figure 2 shows the Al₂O₃-Ti₂O₃-MgO ternary phase diagram at 1873 K. An obvious spinel-stable region can be seen in the diagram, mainly comprising two composite oxide phases:

MgAl₂O₄ and MgTi₂O₄. Analysis of binary and multivariate phase diagrams shows that Mg is key to promoting the formation of composite oxides in the system. Adding Mg causes Al₂O₃ to transform into MgAl₂O₃ and significantly enlarges the stable region of spinel oxides. Therefore, the dominant evolution path of inclusions predicted by thermodynamic calculations is as follows: Al₂O₃ → MgAl₂O₄ → MgAl₂O₄-TiO_x.

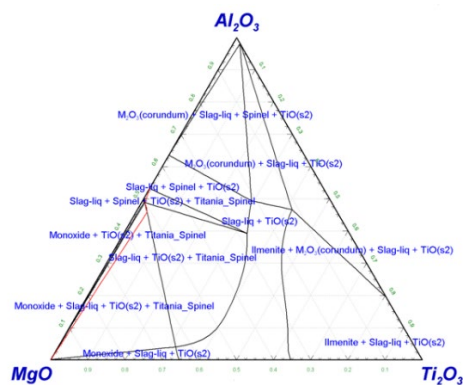


Fig. 2. Ternary phase diagram of the Al₂O₃-Ti₂O₃-MgO system at 1873 K

The thermodynamic calculations above demonstrate that Mg is essential for the formation of composite oxides in the Al-Mg-Ti-O system. However, the mechanism by which Ti incorporates into the Mg-Al-O system warrants further discussion. Based on the present thermodynamic analysis and the literature, Ti can enter the system through two possible pathways.

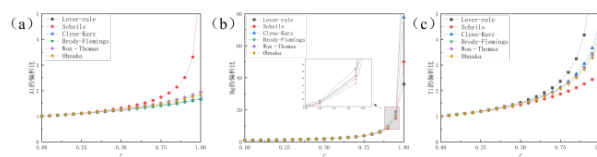
First, from a thermodynamic and crystal-chemical perspective, Ti can incorporate into the MgAl₂O₄ spinel lattice via substitutional solid solution. The spinel structure (AB₂O₄) can accommodate cations of different valences and ionic radii. Ren et al. [23] established the stability phase diagram of Al-Mg-Ti-O inclusions at 1873 K, demonstrating that the stable phases (MgO, Al₂O₃, TiO_x, MgTi₂O₄, and MgAl₂O₄) depend critically on the relative activities of [Al], [Mg], and [Ti] in the molten steel. Specifically, Ti⁴⁺ and Mg²⁺ ions can substitute for Al³⁺ in the MgAl₂O₄ lattice, forming heterovalent substitutional solid solutions that induce lattice distortion and generate cation vacancies. This reduces the lattice energy and promotes further Ti enrichment on the inclusion surface. The presence of Mg enlarges the stability region of spinel oxides (as seen in Fig. 2), creating favorable conditions for Ti to enter the composite oxide system.

Second, from a kinetic perspective, Ti can promote the dissolution of Mg from refractories or slag, as Ti exhibits a high thermodynamic affinity for Mg [24]. The dissolved Mg subsequently reacts with pre-existing Al₂O₃ to form MgAl₂O₄ spinel. Once the MgAl₂O₄ core is formed, Ti-containing species (such as Ti₂O₃ or Ti₂O₅) tend to precipitate on its surface, forming a core-shell structure. This explains why the experimental inclusions observed in Fig. 5 exhibit an Mg-Al-O core with a Ti₂O₃ + MnS outer layer, rather than a homogeneous Mg-Al-Ti-O solid solution. The precipitation sequence is primarily controlled by the segregation behavior of each element: Al₂O₃ precipitates earliest (as shown in Fig. 4a), followed by MgAl₂O₄ formation via modification, and finally Ti oxides precipitate in the late stages of

solidification when the local Ti concentration reaches supersaturation (as shown in Fig. 4b). Thus, the overall evolution pathway is Al₂O₃ → MgAl₂O₄ → MgAl₂O₄ + TiO_x (with MnS), which is consistent with the thermodynamic predictions in Section 3.1 and the experimental observations in Section 3.2.

3.2. Segregation behavior of Al, Mg and Ti elements during solidification

During the solidification of molten steel, the solubility of solute elements in the solid and liquid phases differs, leading to continuous changes in the composition of the residual liquid phase between the dendrites. Figure 3 shows how the interdendritic enrichment factor of the Al, Mg and Ti elements varies with the solid fraction (fs) when the microsegregation model is used to make calculations at a cooling rate of 10 K/min.



(a) Al ; (b) Mg ; (c) Ti

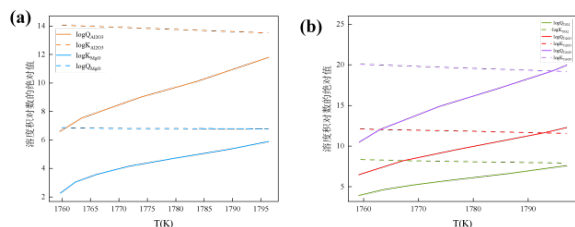
Fig. 3 Segregation degrees of different elements calculated by different models at a cooling rate of 10 K/min

As can be seen in Fig. 3, the three elements become enriched in the residual liquid phase between the dendrites during solidification. The calculation results of the different segregation models are similar in the early and middle stages of solidification, but differ significantly in the late stage. The Scheil model is prone to rapid divergence of solute concentration when fs is close to 1 due to the fact that it ignores solid-phase diffusion. The Lever-rule model assumes that the two-phase solid-liquid is fully diffused and therefore the predicted segregation degree is relatively weak. The Won-Thomas model considers opposite diffusion of the finite solid and can therefore more reasonably describe solute redistribution behaviour in the actual solidification process of molten steel.

In terms of elemental differences, Al shows relatively weak segregation, while Mg and Ti exhibit clear interdendritic enrichment behaviour. Although Mg's distribution coefficient is low, its initial content is small and the absolute concentration is limited. Ti shows significant enrichment during solidification, which is a key factor in the precipitation of Ti-containing oxides in the later stages of solidification.

The effect of different cooling rates on interdendritic enrichment behaviour was analysed further. Cooling rates change the local solidification time and the degree of solid-phase opposite diffusion. When the cooling rate is high, the solidification time is shortened and the reverse diffusion of solute elements in the solid phase is limited. This results in more concentrated solute enrichment in the residual liquid phase between dendrites. Conversely, when the cooling rate is low, the local solidification time is prolonged, solid-phase opposite diffusion is enhanced, and the element distribution is fully adjusted, resulting in relatively moderate end segregation.

Substituting the local liquid phase composition calculated by Eq. (1) into the oxide solubility product criterion, the precipitation conditions of MgO, Al₂O₃, Ti₃O₅ and Ti₂O₃ at different solid fraction can be analyzed.



(a) MgO, Al₂O₃ ; (b) TiO₂, Ti₂O₃, Ti₃O₅

Fig. 4. Variation in the logarithms of actual and theoretical solubility products of MgO, Al₂O₃ and Ti_xO_y oxides with solid fraction

The calculation results show that Al₂O₃ can reach the precipitation condition at the initial stage of solidification, while the precipitation conditions of Ti₃O₅ and Ti₂O₃ are similar. When f_s is between 0.22 and 0.31, the corresponding temperature is approximately 1795 K to 1793 K.

According to the standard formation free energy, the driving force for the formation of MgO is greater than that for the formation of Al₂O₃. However, as the initial Al content in the molten steel is much higher than the Mg content, and Mg activity in the molten steel is low, it is Al₂O₃ rather than MgO that actually reaches the precipitation condition first. As solidification proceeds, the concentration of Mg in the local liquid phase increases and a modification reaction occurs with existing Al₂O₃ inclusions to form spinel-structure inclusions.

In order to verify the calculation results, the industrial production steel EH420 was melted by using the continuous casting slab solidification thermal simulator according to the simulated optimal intercooling speed (50K / min). The samples were observed and analyzed by scanning electron microscope. The results are shown in the figure : The composite inclusions are irregular polygons. The core is composed of elliptical Mg-Al-O spinel inclusions, and Ti₂O₃ and MnS are attached to the periphery.

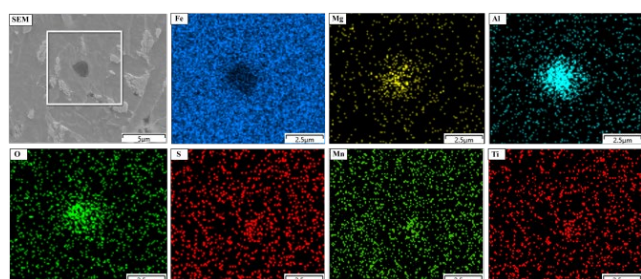


Fig.5 Scanning electron microscope inclusion image and element distribution

4. Conclusions

- 1) Mg is the key element for the transformation of Al₂O₃ to MgAl₂O₄ and Ti composite oxides. The evolution path of inclusions is: Al₂O₃ → MgAl₂O₄ → MgAl₂O₄+ TiO_x cannot form stable composite oxides with Al₂O₃ alone, and Mg must be involved.
- 2) During the solidification process, Al, Mg and Ti are enriched in the residual liquid phase between dendrites, and the segregation of Mg and Ti is more significant. The increase of cooling rate can increase the solute enrichment and promote the precipitation of Ti oxides in the later stage of solidification. The Won-Thomas model can describe this behavior more reasonably due to the consideration of limited solid phase diffusion.
- 3) The precipitation conditions of Al₂O₃ are satisfied at the initial stage of solidification, and the precipitation conditions of Ti₃O₅ and Ti₂O₃ are reached when the solid fraction $f_{\Omega} \approx 0.22 \sim 0.31$ (corresponding to about 1795 ~ 1793 K). The experimental results show that the inclusions have a composite structure of Mg-Al-O core and Ti₂O₃ + MnS shell, which is consistent with the model prediction.
- 4) Mg acts as an essential "bridge" element that enables Ti to enter the Al-O system. Stable Al-Ti oxides cannot form directly; Mg first converts Al₂O₃ into MgAl₂O₄ spinel, which subsequently provides favorable sites for Ti incorporation via substitutional solid solution (Ti⁴⁺ replacing Al³⁺) and surface precipitation.
- 5) The precipitation of Al, Mg, and Ti oxides is not simultaneous but follows a strict thermodynamic sequence. Al₂O₃ precipitates first due to its high initial content, while Ti-oxides reach supersaturation only in the later solidification stage, leading to the observed core-shell structure (MgAl₂O₄ core + TiO_x/ MnS shell).
- 6) Increasing the cooling rate intensifies solute segregation by suppressing solid-state back-diffusion, thereby promoting Ti-oxide precipitation in the final solidification stage. This provides a kinetic route to control the shell thickness and dispersion of complex inclusions.

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