





Phase Transition of Peritectic Steel Q345 and Its Effect on the Equilibrium Partition Coefficients of Solutes

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Received: 16 June 2017; Accepted: 23 July 2017; Published: 28 July 2017

Abstract: The solidification path of peritectic steel Q345 was calculated and compared with in-situ observations to investigate the effect of phase transition on the equilibrium partition coefficient. Subsequently, a thermodynamic model for calculating the equilibrium partition coefficient was established and thermodynamic calculations were performed under different phase configurations. Results indicate that L (liquid phase) + δ , L + δ + γ , and L + δ phases coexist in sequence during the solidification of peritectic steel Q345. The phase constitution of the mushy zone evidently affects the evolution of the equilibrium partition coefficient was quantified through the regression analyses of C, Si, Mn, P, and S solutes under different phase configurations. The average equilibrium partition coefficients of Mn, Si, P, C, and S are 0.696, 0.615, 0.273, 0.2, and 0.033, respectively, thereby indicating the strongest segregation tendency for S and the weakest for Mn.

Keywords: in-situ observation; phase transition; laser scanning confocal microscope (LSCM); equilibrium partition coefficient; thermodynamic calculation

1. Introduction

The equilibrium partition coefficient describes the ratio of solute concentration in the solid and liquid phases during equilibrium solidification [1]. This coefficient plays an important role in segregation problems during alloy solidification [2–4]. Generally, when the partition coefficient is high, the partitioning effect is weak, and consequently, a small segregation degree is obtained during solidification [1]. The equilibrium partition coefficient varies from 0 to 1 for most solutes in steel [2,5], thereby indicating that a solute may be rejected from the solid phase to the liquid phase, which enriches the liquid phase during solidification. The solutes may concentrate in the residual liquid pool due to convection, which results in center segregation at the end of the solidification process of casting steel blank [6–9]. When the partition coefficient is low, segregation is serious.

The equilibrium partition coefficient of solutes in steel highly depends on the phase constitution of the mushy zone, the temperature, and the alloy components [10,11]. To date, some of papers have reported on the evolution of the equilibrium partition coefficient via experiment [1,12] or thermodynamic calculation [10,13]. However, phase transition was not considered in these studies. Moreover, available data on the equilibrium partition coefficient of solutes in peritectic steel remain scarce. For peritectic steel, the solidification path will undergo difference phases of constitution of the mushy zone before and after peritectic reaction. Also, the evolution of the equilibrium partition coefficient during the solidification of peritectic steels is still not clear.

The equilibrium partition coefficient affects solute redistribution in the mushy zone and the segregation degree [4,14]. This coefficient is a critical parameter for segregation simulation and modeling accuracy [15,16]. Fundamental data on the equilibrium partition coefficient of solute

elements for multicomponent steels must be obtained to quantitatively predict segregation during steel solidification. Equilibrium partition coefficients evaluated from binary or ternary systems were frequently used to simulate macrosegregation during the solidification of steel ingot or cast blank due to lack of data [17,18]. Evidently, this approximation may not strictly reflect the true situation of solute redistribution, or the segregation degree in real multicomponent steels (which generally contain more than five different types of solute). Thus, the equilibrium partition coefficient of solutes should be determined within specific multicomponent steels for segregation during solidification by considering phase constitution, temperature, and solute concentration.

In the present work, to get fundamental data on the equilibrium partition coefficient of solute elements for peritectic steel Q345, the solidification path and phase transition of peritectic steel Q345 were studied via phase diagram calculations. Also, the calculated results were compared with the experimental in-situ observations during solidification. Then, the equilibrium partition coefficients were calculated. Phase constitution, temperature, and solute concentration were considered simultaneously. Through the regression analysis, the equilibrium partition coefficients of solutes C, Si, Mn, P, and S in steel Q345 were formulated as a function of temperature under various phase configurations during solidification.

2. Experimental Procedure

The main composition of peritectic steel Q345 is [C] = 0.15, [Si] = 0.27, [Mn] = 1.38, [P] = 0.02, and [S] = 0.009 (in wt %). A high-temperature, laser-scanning confocal microscope (LSCM, VL2000DX, Yonekura MFG Co. Ltd, Osaka, Japan) was used to observe the solidification path and phase transition of peritectic steel in-situ. The schematic diagram of LSCM is shown in Figure 1a. The samples used are machined to a height of 3 mm and a diameter of 7.5 mm. The thermal scheme is shown in Figure 1b. To clearly observe phase transition in the mushy zone, a relatively low cooling rate of 0.3 K/s was adopted within the temperature range from $T_L + 20$ K to $T_S - 20$ K. T_L and T_S are the liquidus and solidus of peritectic steels Q345, respectively.



Figure 1. (a) Schematic diagram of laser scanning confocal microscope (LSCM); (b) the thermal scheme.

3. Thermodynamic Model for the Equilibrium Partition Coefficient

3.1. Model Description

The equilibrium partition coefficient is defined as:

$$k_i = \frac{C_{S,i}^*}{C_{L,i}^*},$$
 (1)

where $c_{S,i}^*$ and $c_{L,i}^*$ are the concentration (wt %) of solute *i* in the solid and liquid phases, respectively, at the solidification interface. When the solidification process is in the thermodynamic equilibrium state, the chemical potential of solute *i* in the solid phase is identical to that in the liquid phase. That is,

$$\mu_{i,0}^{S} + RT \ln a_{i}^{S} = \mu_{i,0}^{L} + RT \ln a_{i}^{L}, \qquad (2)$$

where $\mu_{i,0}^S$ and $\mu_{i,0}^L$ (J/mol) are the chemical potential of solute *i* in the solid and liquid phases, respectively, in the standard state; *R* (J/mol/K) is the gas constant; and *T* (K) is temperature; a_i^S and a_i^L are the activity of solute *i* in the solid and liquid phases, respectively. The relations between a_i and c_i is

$$a_i = \gamma_i c_i. \tag{3}$$

To consider the effect of solute interactions on the equilibrium partition coefficient, the applied activity coefficient γ_i is expressed as:

$$ln\gamma_i = ln\gamma_i^0 + \varepsilon_i^i c_i + \varepsilon_j^j c_j, \tag{4}$$

where γ_i^0 is the activity coefficient during the infinite dilution of solute *i*, ε_i^i is the self-interaction of solute *i*, and ε_i^j is the interaction coefficient between solutes *i* and *j*. Equations (1)–(4) are used to write the thermodynamic formalism of the equilibrium partition coefficient of solute *i* as follows:

$$k_{i}^{0} = \frac{\gamma_{i}^{0,L}}{\gamma_{i}^{0,S}} exp\left[\frac{\mu_{i,0}^{L} - \mu_{i,0}^{S}}{RT} + (\varepsilon_{i}^{i,L}c_{i}^{L} - \varepsilon_{i}^{i,S}c_{i}^{S}) + (\varepsilon_{i}^{j,L}c_{j}^{L} - \varepsilon_{i}^{j,S}c_{j}^{S})\right],$$
(5)

where $\varepsilon_i^{i,S}$ and $\varepsilon_i^{i,L}$ are the self-interaction coefficient of solute *i* in the solid and liquid phases, respectively.

On the right side of Equation (5), the polynomial $(\varepsilon_i^{j,L}c_j^L - \varepsilon_i^{j,S}c_j^S)$ is defined as an effective term, denoting the effect of solute *j* on the equilibrium partition coefficient of solute *i* in the system. From Equation (5), the equilibrium partition coefficient of solute *i* in ferrite (δ) and austenite (γ) can be specified as

$$k_i^{0,\delta} = \frac{\gamma_i^{0,L}}{\gamma_i^{0,\delta}} \exp\left[\frac{\mu_{i,0}^L - \mu_{i,0}^\delta}{RT} + (\varepsilon_i^{i,L}c_i^L - \varepsilon_i^{i,\delta}c_i^\delta) + (\varepsilon_i^{j,L}c_j^L - \varepsilon_i^{j,\delta}c_j^\delta)\right],\tag{6}$$

$$k_i^{0,\gamma} = \frac{\gamma_i^{0,L}}{\gamma_i^{0,\gamma}} exp\left[\frac{\mu_{i,0}^L - \mu_{i,0}^{\gamma}}{RT} + (\varepsilon_i^{i,L}c_i^L - \varepsilon_i^{i,\gamma}c_i^{\gamma}) + (\varepsilon_i^{j,L}c_j^L - \varepsilon_i^{j,\gamma}c_j^{\gamma})\right].$$
(7)

In the L + δ + γ coexistence zone, the equilibrium partition coefficient of solute *i* among δ , γ , and L ($k_i^{0,\delta+\gamma}$) is calculated via

$$k_i^{0,\delta+\gamma} = \frac{m_\delta}{m_\delta + m_\gamma} \times k_i^{0,\delta} + \frac{m_\gamma}{m_\delta + m_\gamma} \times k_i^{0,\gamma},\tag{8}$$

where m_{δ} and m_{γ} are the mass of δ and γ phases, respectively, during the solidification process. For a multicomponent system, the effect of solutes on the equilibrium partition coefficient was considered by introducing effective terms into the model. In the current study, all calculations were performed using the Equilib module of FactSage 6.3, in which the database FSstel is selected [19].

3.2. Model Verification

To verify the accuracy of the present model, the equilibrium partition coefficients of C in the Fe-0.93%C-1%Mn ternary system and the Fe-1.27%C-2.03%Si-0.88%Mn quaternary system were calculated, and the results were compared with that from References [12,13]. As shown in Figure 2, the calculation results of the present model for the ternary Fe-0.93%C-1%Mn system are in accordance

with the calculation results from Reference [13]. The predicted average k_C in the entire mushy zone is 0.350, which is extremely close to the value of 0.352 from Reference [13]. The deviation between our prediction and the literature result reaches as low as 0.57%. For the quaternary Fe-1.27%C-2.03%Si-0.88%Mn system, k_C from the present model exhibits the same tendency as the experimental data presented in Reference [12]. The average k_C calculated from our model is 0.396, whereas the experimental value is 0.421, which presents a small negative deviation (i.e., 5.94%). Evidently, the results from the present model demonstrate relatively high reliability with respect to reference data. Thus, the model was used for the subsequent thermodynamic calculations.



Figure 2. Comparison of k_c between the present calculated results and those of the literature (Data of Kagawa et al. from Reference [13]; Data of Ocansey et al. from Reference [12]).

4. Results and Discussion

4.1. Solidification Path and Phase Transition of Peritectic Steel Q345

The Fe-C-0.27%Si-1.38%Mn-0.02%P-0.009%S phase diagram of steel Q345 was calculated and shown in Figure 3a. The diagram illustrates that $L + \delta$, $L + \delta + \gamma$, and $L + \gamma$ phases coexist in sequence in the solidification path of steel Q345. From 1786 K to 1755 K, L gradually transforms into the δ phase while the L and δ phases coexist. L is residual after the precipitation of δ . A peritectic reaction occurs from 1755 K to 1753 K. The residual L and δ transform to γ until δ is exhausted. L, δ , and γ coexist within this temperature range. From 1753 K to 1737 K, γ will directly precipitate from the residual L while L and γ coexist. When temperature decreases to 1737 K, steel Q345 solidifies completely. An MnS inclusion is not formed in the mushy zone during the solidification of steel Q345 due to the low concentration of Mn and S; this finding is consistent with the report by Bao [20]. Therefore, the effect of MnS precipitation on the equilibrium partition coefficients of Mn and S should no longer be considered. In addition, the redistribution of solutes during the solidification process is illustrated in Figure 3b. This figure indicates that the solutes are rejected from the solid phase to the liquid phase (as the black arrows shown) during solidification, thereby enriching the liquid phase. Firstly, solutes are redistributed between δ and L in the L + δ coexistence zone. Secondly, solutes are redistributed among δ , γ , and L in the L + δ + γ coexistence zone with the formation of γ . As temperature decreases, the δ phase disappears and solutes are redistributed between γ and L in the L + γ coexistence zone. Thus, the equilibrium partition coefficients should be determined quantitatively according to the phase constitution of the mushy zone during solidification.



Figure 3. (a) Solidification path of steel Q345 and (b) the solutes redistribution process.

The in-situ observations of the phase transition in the mushy zone of steel Q345 are shown in Figure 4, which indicates that the phase transition undergoes three stages. Firstly, L transforms into δ and the grains grow with decreasing temperature, as shown in Figure 4a–c. Secondly, the peritectic reaction occurs, i.e., L and δ transform into γ . L is residual after the peritectic reaction. This process is shown in Figure 4d–f. Third, residual L gradually transforms into γ until complete solidification is achieved, as shown in Figure 4g–i. The in-situ observations of phase transitions confirm the accuracy of the thermodynamic calculations for the solidification path of peritectic steel Q345.



Figure 4. Cont.



Figure 4. In-situ observation of phase transition in the mushy zone of steel Q345; from (**a**) to (**c**): L transforms into δ ; from (**d**) to (**f**): L and δ transform into γ ; from (**g**) to (**i**): L transforms into γ .

4.2. Evolution of Equilibrium Partition Coefficients during Solidification and Phase Transition

The evolution of k_C during the solidification of steel Q345 is shown in Figure 5a. The figure indicates that the equilibrium partition coefficient of C between the δ and L phases (k_C^{δ}) is higher than that between the γ and L phases (k_C^{γ}). At temperatures ranging from 1786 K to 1755 K, k_C^{δ} is nearly a constant with a value of 0.148.

When temperature falls below 1755 K, the γ phase forms from the peritectic reaction $(L + \delta \rightarrow \gamma)$. The equilibrium partition coefficient of C in the L + δ + γ coexistence zone $(k_C^{\delta+\gamma})$ rapidly increases with an increasing amount of γ because k_C^{γ} is nearly twice k_C^{δ} at the same temperature.

When the peritectic reaction $(L + \delta \rightarrow \gamma)$ terminates at 1753 K, the δ phase disappears and L and γ coexist in the mushy zone. In the coexistence zone, i.e., from 1753 K to 1737 K, k_C^{γ} further increases with decreasing temperature.

The evolution of k_C varies with phase configurations; hence, the temperature dependence of k_C for steel Q345 is obtained via regression analysis according to phase transitions; that is,

$$k_{\rm C} = \begin{cases} 0.148 & 1755 \,\,{\rm K} < T \le 1786 \,\,{\rm K} \\ -0.03825T + 67.33547 & 1753 \,\,{\rm K} \le T \le 1755 \,\,{\rm K} \\ -6.80795 \times 10^{-4}T + 1.48018 & 1737 \,\,{\rm K} \le T < 1753 \,\,{\rm K} \end{cases}$$
(9)

The evolution of the equilibrium partition coefficients of Si, Mn, P, and S during the solidification of steel Q345 are shown in Figure 5b–e, respectively. k_{Si}^{δ} increases with decreasing temperature. From the L + δ zone to the L + δ + γ zone, $k_{Si}^{\delta+\gamma}$ decreases marginally because k_{Si}^{γ} is slightly lower than k_{Si}^{δ} [2]. The effect of the peritectic reaction on the equilibrium partition coefficient of Si is negligible. k_{Si}^{γ} increases with decreasing temperature. When temperature decreases from 1786 K to 1737 K, k_{Si}^{γ} increases from 0.579 to 0.667, i.e., an increase of 15.2%. Thus, the effect of temperature on the equilibrium partition coefficient of Si is not ignorable.

 k_{Mn}^{δ} is reduced with decreasing temperature. From the L + δ zone to the L + δ + γ zone, $k_{Mn}^{\delta+\gamma}$ rapidly increases because k_{Mn}^{γ} is greater than k_{Mn}^{δ} [2]. In addition, the effect of phase constitution in the mushy zone on k_{Mn} is more significant than that of temperature. In the L + γ coexistence zone, k_{Mn}^{γ} gradually decreases with temperature.

The equilibrium partition coefficients of P and S exhibit similar variation trends. In the L + δ coexistence zone, k_P^{δ} and k_S^{δ} slightly increase with decreasing temperature. From the L + δ zone to the L + δ + γ zone, $k_P^{\delta+\gamma}$ and $k_S^{\delta+\gamma}$ decreases sharply due to γ phase formation from the peritectic reaction. In the L + γ coexistence zone, k_P^{γ} is nearly constant, whereas k_S^{γ} declines with decreasing temperature.

k

The temperature dependence of k_{Si} , k_{Mn} , k_P , and k_S in the three coexistence zones during the solidification of steel Q345 is obtained from Equations (10)–(13).

$$k_{Si} = 1.50975 \times 10^{-5} T^2 - 0.05492T + 50.5077$$
 1737 K $\leq T \leq 1786$ K, (10)

$$k_{Mn} = \begin{cases} 0.00102T - 1.12182 & 1755 \text{ K} < T \le 1786 \text{ K} \\ -0.01264T + 22.87046 & 1753 \text{ K} \le T \le 1755 \text{ K} \\ -0.00102T - 1.06985 & 1737 \text{ K} \le T < 1753 \text{ K} \end{cases}$$
(11)

$$P_{P} = \begin{cases} -3.51091 \times 10^{-4} T - 0.95203 & 1755 \text{ K} < T \le 1786 \text{ K} \\ 0.04538T - 79.37208 & 1753 \text{ K} \le T \le 1755 \text{ K} \\ 0.170 & 1737 \text{ K} \le T < 1753 \text{ K} \end{cases}$$
(12)

$$k_{S} = \begin{cases} -2.10638 \times 10^{-5}T - 0.95203 & 1755 \text{ K} < T \le 1786 \text{ K} \\ 0.058T - 10.15078 & 1753 \text{ K} \le T \le 1755 \text{ K} \\ 8.66261 \times 10^{-5}T - 0.13217 & 1737 \text{ K} < T < 1753 \text{ K} \end{cases}$$
(13)

The average equilibrium partition coefficients were analyzed and illustrated in Figure 5f. As shown in the Figure 5f, ave_ks is the minimum, thereby indicating that S in steel Q345 exhibits an ultra-strong tendency toward segregation, which is consistent with Reference [4]. ave_kc is 0.2, which is slightly higher than that of S, is followed by P, with an ave_kp of 0.273. Similar to S, C and P demonstrate a relatively strong segregation tendency. ave_ks_i and ave_km_n are equal to 0.615 and 0.696, respectively. That is, the solute sequence arranged in order of weakening segregation tendency in steel Q345 is S, C, P, Si, and Mn.



Figure 5. Evalution of (a) k_C , (b) k_{Si} , (c) k_{Mn} , (d) k_P , (e) k_S , and (f) average equilibrium partition coefficients of solutes during peritectic steel Q345 solidification.

5. Conclusions

- (1) The L + δ , L + δ + γ , and L + γ phases coexist in sequence in the solidification path of steel Q345, which agrees well with in-situ observations.
- (2) The temperature dependent equilibrium partition coefficients of the solutes in Q345 were quantitatively studied, and regression formulas were established under various phase constitutions.
- (3) The solute sequence arranged in order of weakening segregation tendency in steel Q345 is S, C, P, Si, and Mn. The average equilibrium partition coefficients of these constituents are 0.696, 0.615, 0.273, 0.2, and 0.033, respectively.

Acknowledgments: The authors would like to thank the Natural Science Foundation of China (NSFC) for the financial support (Project Nos.: 51504048; 51374260; 51611130062).

Author Contributions: Huabiao Chen, Mujun Long, and Dengfu Chen conceived and designed the experiments; Huabiao Chen, Junsheng Cao, ZhiHua Dong, and Tao Liu performed the experiments and calculations; All of the authors analyzed and discussed the data; Huabiao Chen wrote the paper.

Conflicts of Interest: The authors declare no conflict of interest.

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