



Study on Mn Volatilization Behavior During Vacuum Melting of High-manganese Steel

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Abstract

As an alloying element in steel, manganese can considerably enhance the mechanical properties of structural steel. However, the Mn volatilisation loss in vacuum melting is severe because of the high saturated vapour pressure, resulting in an unstable Mn yield and Mn content fluctuation. Therefore, a systematic study of the volatilisation behaviour of Mn in vacuum melting is required to obtain a suitable Mn control process to achieve precise control of Mn composition, thereby providing a theoretical basis for industrial melting of high-Mn steel. In order to explore the Mn volatilization behavior, the volatilization thermodynamics and volatilisation rate of Mn, as well as the influence factors are discussed in this study. The results shows that Mn is extremely volatilised into the vapour phase under vacuum, the equilibrium partial pressure is closely related to Mn content and temperature. With an increase in the Mn content, a higher C content has a more obvious inhibitory effect on the equilibrium partial pressure of Mn. The maximum theoretical volatilisation rate of Mn shows a linear upward trend with an increase in Mn content. However, a higher C content has a more obvious effect on the reduction of the maximum theoretical volatilisation rate with the increase of Mn content. This study provides an improved understanding of Mn volatilisation behaviour as well as a theoretical foundation for consistent Mn yield control during the vacuum melting process of high-Mn steel.

Keywords: Mn volatilization behavior, Vacuum melting, Thermodynamics, High-manganese steel

1. Introduction

As is well known, manganese (Mn) as an alloying element in steel can significantly improve the mechanical properties of structural steel. In the steel industry, 70% of Mn is added to steel alloying, which improves the properties of steel. The automobile industry is the main user of the steel industry, and ultrahigh-strength steel is particularly used in the automobile industry due to its good formability and high mechanical properties of strong plasticity contributes to the lightweight design of automobiles [1–3]. In modern automobile manufacturing, the steel proportion is usually greater than 45% and is increasing yearly. To make automotive steel have a higher lightweight potential, ultrahigh-strength steel is strengthened by solid solution strengthening,

precipitation strengthening and grain refining of alloying elements. Mechanisms such as transformation-induced plasticity (TRIP) and twinning-induced plasticity (TWIP) are also employed to improve plasticity and formability [4–6]. Therefore, the TWIP/TRIP steel with 15 wt%–33 wt% Mn content has become the main development direction of next-generation automotive steel [7, 8]. Grässel et al. found that when the Mn content of Fe–Mn–Si–Al series TRIP steel reached 25 wt%, the strong plastic volume reached 50 GPa% [9]. The tensile strength of Fe–25Mn–3Si–3Al series TWIP steel is 650 MPa because of the formation of twins at various strain rates [10]. However, the formation of twins depends on the austenite layer fault energy, which is mainly related to the chemical composition of Mn-containing high-strength steel [11–13].



At present, the research and development of ultrahigh-strength high-Mn TWIP steel have been actively conducted. Due to the advantages of low segregation, high cleanliness, dense structure and good mechanical properties for vacuum melting technology, vacuum melting has become crucial in the melting process of high-end metal materials, and is often used in melting high-quality special steel [14, 15]. However, the Mn volatilisation loss in vacuum melting is severe because of the high saturated vapour pressure [16], resulting in an unstable Mn yield and Mn content fluctuation. At present, the composition of the base metal is determined only by experience. Therefore, to achieve high-quality melting of high-Mn steel, precise control of Mn composition is crucial. While, there are only a few studies on the control of Mn volatilisation in traditional steel with low or medium Mn content [17, 18], and the control of inclusion for medium/high-Mn steel. There are few reports on the control of Mn volatilisation in high-Mn steel under vacuum condition. The volatilisation behaviour of high-Mn steel under high temperature and vacuum determines the loss of Mn and the stability of Mn composition control in steel. Therefore, it is necessary to systematically study the volatilisation behaviour of Mn in the vacuum melting process and obtain a suitable Mn control process to achieve precise control of Mn content.

In this study, the Mn volatilization behavior and its influencing factors are discussed. This study offers an improved understanding of the volatilisation behaviour of Mn and a theoretical basis for the stable control of Mn yield during the vacuum melting process of high-Mn steel.

2. Methodology

For the study the chemical composition of the high-Mn steel is 0.20 wt% C, 25 wt% Mn, 3.0 wt% Si, 3.0 wt% Al, 0.01 wt% P, 0.007 wt% S, 0.0015 wt% O, 0.0025 wt% N, and balance Fe, respectively. The contents O and N were analyzed by a oxygen-nitrogen analyzer, the contents of C and S were analyzed by a carbon-sulfur analyzer, and the content of other elements were analyzed by an optical emission spectrometer. The volatilization behavior of Mn is analyzed in terms of volatilization thermodynamics and volatilization rate. Herein, the volatilization thermodynamics is analyzed according to the saturation vapour pressure and Gibbs free energy based on the thermodynamic theory of high temperature metal solution, and the critical pressure combined with the statistical physics approach is applied to explore the maximum theoretical volatilisation rate. Meanwhile, the influence factors of temperature, manganese content, and carbon content are discussed according to the actual vacuum melting parameters for the high-Mn steel.

3. Results and Discussion

3.1. Thermodynamics of Mn Volatilisation

The fundamental physical volatilisation of metal elements from steel can occur in a specific temperature range, which is based on

the principle that the saturation vapour pressure significantly contributes to the evaporation characteristics. In general, the higher the saturated vapour pressure of an element, the easier it is to volatilise it. According to the Clausius-Clapeyron relation, the saturation vapour pressures of Mn, Al, Fe and Si can be, respectively, calculated as follows [19]:

$$\log p_{\text{Mn}}^* = -\frac{14520}{T} - 3.02\log T + 21.36 \quad (1)$$

$$\log p_{\text{Al}}^* = -\frac{16450}{T} - 1.02\log T + 14.48 \quad (2)$$

$$\log p_{\text{Fe}}^* = -\frac{19710}{T} - 1.27\log T + 15.39 \quad (3)$$

$$\log p_{\text{Si}}^* = -\frac{20900}{T} - 0.57\log T + 12.9 \quad (4)$$

where p^* denotes the saturated vapour pressure of the pure component (Pa) and T denotes the absolute temperature (K). The relationships between the saturated vapour pressure and temperature of Mn, Al, Fe and Si are plotted in Fig. 1.

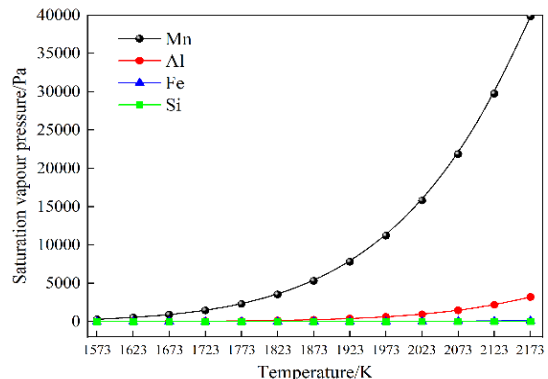


Fig. 1. The saturation vapour pressures of Mn, Al, Fe and Si

Fig. 1 shows that the saturated vapour pressure of Mn increases exponentially with the increase of temperature. At the same temperature, the saturated vapour pressure of Mn is significantly higher than those of Al, Fe and Si. When the temperature is 1873 K, the saturated vapour pressure of Mn reaches up to 5370 Pa, indicating that Mn is extremely volatilised into the vapour phase under vacuum. Therefore, at the conventional liquid steel refining temperature (1873 K) and pressure (67 Pa), the evaporation loss of Al, Fe and Si in liquid steel are negligible. Thus, the volatilisation behaviour of Mn is mainly discussed.

The transformation of dissolved Mn into gaseous Mn in liquid steel must meet certain thermodynamic conditions. Its gasification reaction and Gibbs free energy in the standard state are, respectively, given by as follows [20]:

$$[\text{Mn}] = \text{Mn}(\text{gas}) \quad (5)$$

$$\Delta G_{\text{Mn}}^{\theta} = -231720 - 63.01T \quad (6)$$

The Gibbs free energy of the reaction in the non-standard state is given by the following equation:

$$\Delta_r G_{Mn} = \Delta G_{Mn}^\theta + RT \ln \left(\frac{p_{Mn}/p^\theta}{f_{Mn} w_{Mn}} \right) \quad (7)$$

where R denotes the ideal gas constant (8.314 J/mol/K), p_{Mn} denotes the vapour partial pressure of Mn (Pa), and f_{Mn} and w_{Mn} denote the activity coefficient and mass fraction of Mn in liquid steel, respectively. The activity coefficient of Mn in liquid steel is related to the content of other elements, which can be calculated according to Wagner's equation, as follows [21]:

$$\log f_{Mn}(1873 \text{ K}) = \sum e_{Mn}^j [\%j] \quad (8)$$

where e_{Mn}^j denotes the interaction coefficient of element j on Mn in steel at 1873 K [22], as listed in Table 1.

Table 1.
Interaction coefficients in molten steel at 1873 K

e_{Mn}^j	C	Si	Mn	Al	P	S	O	N
Mn	-0.07	-0.0002	0	0.07	-0.0035	-0.048	-0.083	-0.091

The effect of temperature on activity coefficient f_{Mn} is given by the following equation [23]:

$$\log f_{Mn}(T) = \left(\frac{2538}{T} - 0.355 \right) \log f_{Mn}(1873 \text{ K}) \quad (9)$$

When the reaction reaches equilibrium $\Delta_r G_{Mn} = 0$, the following relationship can be obtained from the above equation:

$$\frac{231720}{T} = 63.01 - 19.143 \left[\log \left(\frac{p_{Mn}}{p^\theta} \right) - \log f_{Mn} - \log w_{Mn} \right] \quad (10)$$

3.2. Influencing Factors on Partial Pressure

The relationship between the temperature of molten steel and the equilibrium partial pressure of Mn can be obtained from Equation (10), as shown in Fig. 2. From the figure, the equilibrium partial pressure of Mn increases with Mn content, and the higher the temperature, the more significant the rising trend. Under a constant Mn content, the equilibrium partial pressure of Mn is mainly affected by temperature. However, the degree of influence of temperature is closely related to the initial Mn content. The higher the initial Mn content, the greater the influence degree of temperature.

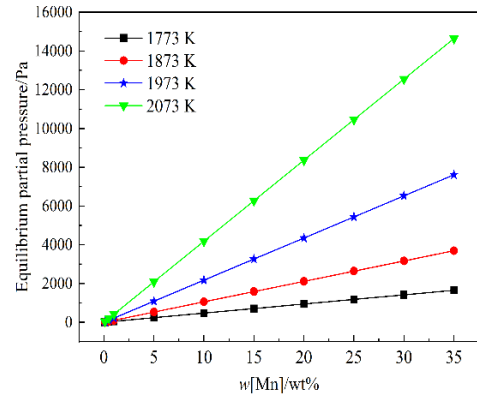


Fig. 2. The effect of Mn on equilibrium partial pressure

Other elements that influence Mn volatilisation in liquid steel are mainly C, Si, Al, P, S, O and N. In general, the content of harmful P, S, O and N elements is low. While there is a large change in the C element content, from approximately 0.1 wt% to 2 wt%. Therefore, the influence of C content on the volatilisation of Mn cannot be ignored. To this end, the effect of C content on the Mn equilibrium partial pressure are further analysed, as showing Fig. 3. It shows the influence of different C contents and temperatures on the equilibrium partial pressure of Mn when the Mn content is 25 wt%. From the figure, the equilibrium partial pressure of Mn decreases with increasing C content, and the higher the temperature, the more obvious the decline. Therefore, for high-Mn steel, the equilibrium partial pressure of Mn is inhibited by an increase in the C content at higher temperatures.

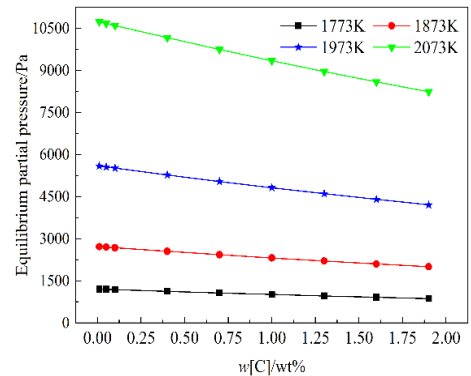


Fig. 3. The effect of C on equilibrium partial pressure

Fig.4 shows the effect of different C and Mn contents on the equilibrium partial pressure of Mn at 1873 K. From the figure, the inhibition effect of the C content on the equilibrium partial pressure of Mn is related to the Mn content. When the Mn content is 1 wt%, the C content has almost no inhibitory effect. With an increase in the Mn content, a higher C content has a more obvious inhibitory effect on the equilibrium partial pressure of Mn.

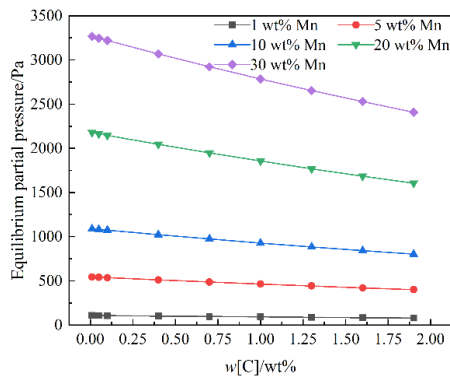


Fig. 4. The effect of C and Mn on equilibrium partial pressure

When Mn begins to volatilise at the conventional melting vacuum of 67 Pa, the corresponding theoretical equilibrium transformation temperature is shown in Fig. 5. The equilibrium transformation temperature decreases with increasing Mn content, which is mainly related to the higher equilibrium partial pressure corresponding to higher Mn content. In addition, when the Mn content is constant, a higher C content increases the equilibrium theoretical transformation temperature of Mn. Whereas, a lower C content, such as 0.01 wt% and 0.10 wt%, has little influence on the equilibrium theoretical transformation temperature.

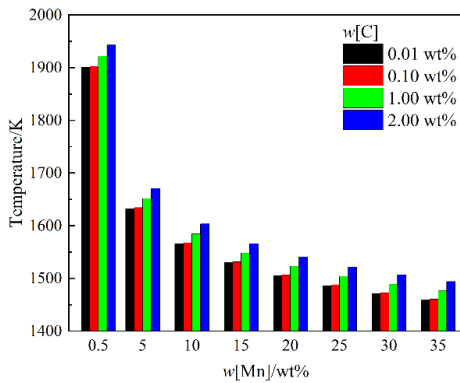


Fig. 5. The effect of C and Mn on theoretical equilibrium transformation temperature

3.3. Analysis of Mn Volatilisation Rate

In general, the rate of volatilisation of metals is closely related to the pressure of a system. According to relevant studies, the volatilisation of metals can be divided into general, explosive and molecular evaporations, and the volatilisation rate is as follows: general < explosive < molecular. When the pressure is further reduced, the volatilisation rate increases significantly. When the volatilisation rate is continuously reduced to a certain pressure, it no longer increases and enters the molecular volatilisation stage, as shown in Fig. 6. At this time, the pressure value is called the critical pressure p_{cri} .

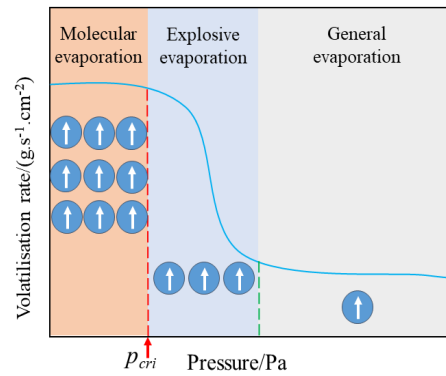


Fig. 6. The different volatilisation stages of metals

The critical pressure can be calculated as follows [24]:

$$p_{cri} = \frac{kT}{\lambda\sigma\sqrt{1+\frac{M_{Mn}}{M_{gas}}}} \quad (11)$$

where k is the Boltzmann constant (6.626×10^{-34} J/s), λ denotes the mean free path of a gas and σ denotes the average effective cross-section of the gas molecules. The molar masses of M_{Mn} and M_{gas} , are 55 g/mol and 29 g/mol, respectively.

According to the statistical physics, the λ and n can be obtained [25]:

$$\lambda = \frac{1}{\sqrt{2}n} \quad (12)$$

$$n = \frac{p_{Mn}^* + p_{res}}{kT} \quad (13)$$

where n is the molecular density of the gas and p_{res} denotes the system residual pressure. Combining the above equations, then the Equation (14) can be obtained:

$$p_{cri} = \frac{\sqrt{2}(p_{Mn}^* + p_{res})}{\sqrt{1+\frac{M_{Mn}}{M_{gas}}}} \quad (14)$$

Because p_{Mn}^* is significantly greater than p_{res} , thus the Equation (14) can be simplified as:

$$p_{cri} = \frac{\sqrt{2}p_{Mn}^*}{\sqrt{1+\frac{M_{Mn}}{M_{gas}}}} \quad (15)$$

Then, the critical pressure of Mn, $p_{Mn,cri}$, is given by follows:

$$p_{Mn,cri} = a_{Mn}p_{cri} = \frac{\sqrt{2}(p_{Mn}^*)a_{Mn}}{\sqrt{1+\frac{M_{Mn}}{M_{gas}}}} \quad (16)$$

$$a_{Mn} = x_{Mn}\gamma_{Mn} = \frac{56}{5500}f_{Mn}w_{Mn} \quad (17)$$

where a_{Mn} denotes the activity based on Raoult's law and x_{Mn} and γ_{Mn} denote the molar fraction of Mn and the Raoult activity coefficient, respectively.

For the volatilisation rate of metals, Hertz and Knudsen modified the Langmuir equation and obtained the expression for the volatilisation rate [26]:

$$v = p_{Mn} \sqrt{\frac{M_{Mn}}{2\pi RT}} \quad (18)$$

When $p_{Mn} = p_{Mn,cri}$, the volatilisation rate of Mn reaches the maximum theoretical value, which can be calculated as follows:

$$v = p_{Mn,cri} \sqrt{\frac{M_{Mn}}{2\pi RT}} \quad (19)$$

3.4. Influencing Factors on Volatilisation Rate

The critical pressure and the maximum theoretical volatilisation rate at different temperatures and Mn contents are shown in Fig. 7. It shows that the critical pressure and the maximum theoretical volatilisation rate present a linear upward trend with an increase in Mn content, and the higher the temperature, the more significant the upward trend. Under a constant Mn content, the critical pressure and the maximum theoretical volatilisation rate are mainly affected by temperature. However, the degree of influence of temperature is related to the Mn content. The higher the Mn content, the more significant the critical pressure and the maximum theoretical volatilisation rate rise. When the temperature is 1873 K, the maximum theoretical volatilisation rate of Mn increases from 1.67 to 58.59 $\text{g}\cdot\text{s}^{-1}\cdot\text{cm}^{-2}$ as the Mn content increases from 1 wt% to 35 wt%.

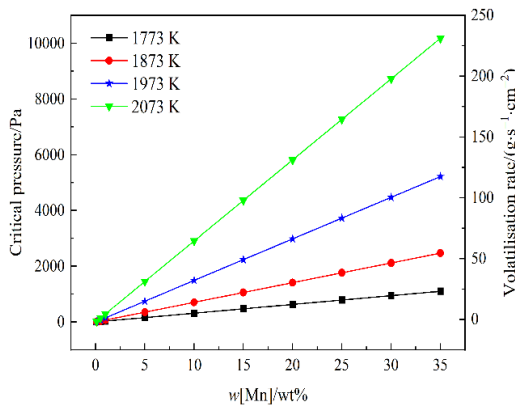


Fig. 7. The effect of Mn on critical pressure and maximum theoretical volatilisation rate

The influence of different C and Mn contents on the critical pressure and the maximum theoretical volatilisation rate of Mn at 1873 K is shown in Fig. 8. It shows that the critical pressure and the maximum theoretical volatilisation rate decrease with increasing C content. However, the decreasing trend is related to the Mn content, when the Mn content is 1 wt%, the C content has almost no effect. While, with an increase in Mn content, a higher C content has a more obvious effect on the reduction of the critical pressure and the maximum theoretical volatilisation rate. When the Mn content is 20 wt% and the C content increases from 0.01 wt%

to 1.9 wt%, the maximum theoretical volatilisation rate decreases from 34.52 to 25.46 $\text{g}\cdot\text{s}^{-1}\cdot\text{cm}^{-2}$.

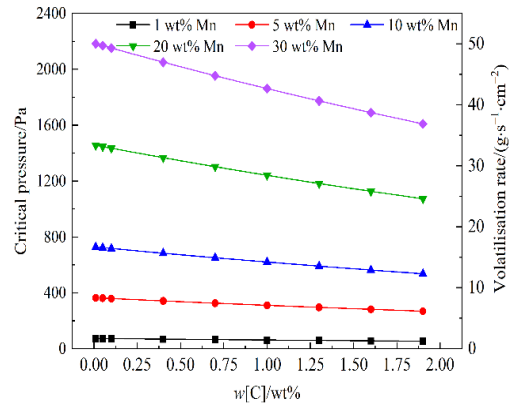


Fig. 8. The effect of C on critical pressure and maximum theoretical volatilisation rate

Then, the corresponding Mn content in molten steel according to the equilibrium partial pressure and the critical pressure of Mn under the conventional melting vacuum (67 Pa) can be calculated and the Mn content region corresponding to the explosive evaporation stage can be obtained, as shown in Fig. 9. When the Mn content exceeds the boundary line where the spherical symbol is located, it will enter the molecular evaporation stage. When the Mn content is lower than the boundary line where the diamond symbol is located, it enters the general evaporation stage. From the Figure 9, the corresponding Mn content region presents an upward trend with an increase in C content, and its change trend is closely related to the temperature. When the temperature is relatively low, the corresponding area is larger and the rising trend is more obvious. With an increase in temperature, the corresponding Mn content region becomes smaller and the rising trend becomes slower. This indicates that Mn easily enters the molecular volatilisation stage, and the C content has almost no obvious inhibitory effect when the temperature is high. Therefore, at the conventional steelmaking temperature and vacuum, the volatilisation of Mn occurs mainly in the molecular evaporation stage of high-Mn steel.

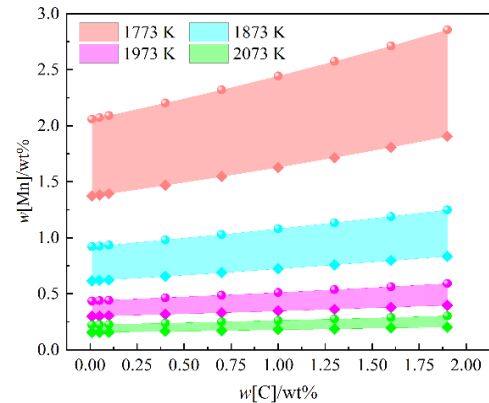


Fig. 9. Mn content region for the explosive evaporation stage

4. Conclusions

Thermodynamic analysis shows that, Mn is extremely volatilised into the vapour phase under vacuum at steelmaking temperature. The equilibrium partial pressure of Mn increases with Mn content and temperature. While with an increase in the Mn content, a higher C content has a more obvious inhibitory effect on the equilibrium partial pressure of Mn.

The maximum theoretical volatilisation rate of Mn shows a linear upward trend with an increase in Mn content. However, a higher C content has a more obvious effect on the reduction of the maximum theoretical volatilisation rate with the increase of Mn content.

The Mn content region corresponding to the explosive evaporation stage under conventional melting vacuum presents an upward trend with an increase in C content, and its change trend is closely related to the temperature.

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