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INVESTIGATION ON IMPROVING THE CASTABILITY OF LOW CARBON AI-KILLED STEEL WITHOUT CALCIUM TREATMENT

During the "BOF-LF-CC" process of producing low carbon Al-killed (LCAK) steel, Al in the molten steel will react with MgO in the ladle refractory or the refining slag to generate MgO-Al₂O₃ inclusions, which have a negative influence on the molten steel's castability. To enhance the castability of molten steel, calcium treatment is typically required following LF refining to promote the transformation of inclusions MgO-Al₂O₃ to CaO-MgO-Al₂O₃ or CaO-Al₂O₃. However, calcium treatment has many drawbacks, such as low calcium yield, increased smelting cost, environmental pollution, etc. Thus, how to improve the castability of LCAK steel without calcium treatment is worth studying. In this research, laboratory studies were first carried out to clarify the source of MgO-Al₂O₃ inclusions in the molten steel. Thereafter, industrial trials were conducted with the refractory material of the ladle replaced by a MgO-free and Al₂O₃-riched refractory. The results show that when a MgO-based crucible is used at 1600°C, the inclusions in molten steel after 25 min are mainly MgO-Al₂O₃, even without refining slag. However, even with the refining slag (the basicity is less than 4.5) containing about 5% MgO, when an Al₂O₃-based crucible is employed, the inclusions in the molten steel are mainly CaO-Al₂O₃. Consequently, MgO in ladle refractories is the main source for the formation of inclusions MgO-Al₂O₃. The results of industrial trials using the "3 + 1" smelting pattern, in which the molten steel is cast directly without calcium treatment in the first three heats, treated with calcium in the next heat, and the process is repeated, show a significant improvement in the castability of molten steel.

Keywords: Low Carbon Al-killed Steel; Aluminum deoxidation; LF refining; Inclusions; Calcium treatment

1. Introduction

There are three main processes for the production of low carbon Al-killed (LCAK) Steel: "BOF-LF-CC", "BOF-RH-CC", and "BOF-CAS-CC" [1]. Because LF refining has the advantages of strong desulphurization ability and stable controlling of temperature, the BOF-LF-CC process is adopted to produce LCAK steel by the vast majority of steel enterprises. For the BOF-LF-CC process, the refining slag with high basicity and low oxidizable is usually used to improve the effect of desulfurization, deoxidation and inclusion control in the LF refining process [2-3]. However, due to the high basicity and low oxygen potential of the refining slag, Eqs. (1)~(3) occur during the refining process [4-7]. MgOžAl₂O₃ inclusions generated by Eqs. (1)~(3) seriously deteriorate the castability of the molten steel [8].

$$3(MgO)_{Slag/Refractory} + 2[Al] = 3[Mg] + + (Al_2O_3)_{Slag/Inclusions}$$
(1)

$$[Mg] + 4[O] + 2[Al] = (MgO \cdot Al_2O_3)_{Inclusion}$$
(2)

$$[Mg] + n/3(Al_2O_3)_{Inclusion} =$$

= (MgO \cdot (n - 1)/3Al_2O_3)_{Inclusion} + 2/3[Al] (3)

Because the contact area between ladle refractory material and molten steel is obviously larger than that between refining slag and molten steel, the impact of ladle refractory material on MgO-Al₂O₃ inclusion generation in molten steel has been getting attention [9]. Brabie [10] found that MgO in ladle refractory materials would be reduced by the carbon in the refractory, causing the content of dissolved [Mg] in molten steel would increase and the formation of MgO-Al₂O₃ inclusion. Chi et al. [11], Liu et al. [12-13] and Jansson et al. [14] investigated the influence of MgO-refractory materials on the inclusion formation in molten steel and achieved comparable experimental results.

Wang et al. [15] studied the mass transfer process among molten steel, slag, lining refractory and non-metallic inclusions in the refining process of bearing steel by combining labora-

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tory experiments with numerical simulation and prediction, and found that refining slag significantly accelerated the mass transfer of inclusions. Liu et al. [9,12-13] compared the effects of MgO-containing refining slag and different MgO-based refractory materials on inclusion transformation in steel through experiments, and the results show that the rates at which MgO refractories, MgO-C refractories and MgO-containing refining slag to supply dissolved magnesium to steel are 2.18×10^{-4} , 5.0×10^{-4} , 6.3×10^{-4} m/s, respectively. Meanwhile, Liu et al. [13] also pointed out that while the MgO-containing slag supplies magnesium at a faster rate than MgO-C and MgO refractory, the impact of ladle refractory material on MgO-Al₂O₃ inclusion in steel is greater than that of the refining slag when taking contact area into account. Deng et al. [7,16] studied the effect of MgO-Al₂O₃ refractory materials on inclusions in Al-deoxidized steel and discovered that the Al₂O₃ inclusions do not transform in molten steel due to low MgO activity in Mg-Al refractory materials and the difficulty of being reduced by [Al] in molten steel.

Itoh et al. [4] found through thermodynamic calculation that MgO-Al₂O₃ inclusions in the molten steel can be transformed into more stable Al₂O₃-MgO-CaO liquid inclusions when the steel contains a certain amount of Ca. This finding proves the feasibility of using calcium treatment to transform MgO-Al₂O₃ inclusions into liquid inclusions from a thermodynamic standpoint. Park et al. [17] indicated that when a CaO-A12O3-MgO slag system with high basicity (>6.0), high calcium and high aluminum is utilized in the smelting of stainless steel, MgO-Al₂O₃ inclusions in steel will transform to Al2O3-MgO-CaO inclusions. Todoroki et al. [18] also studied the viability of modifying MgO-Al₂O₃ inclusions using a refining slag system, but all of the experiments were conducted in the laboratory. Yu et al. [19] found that increasing the basicity of refining slag $(w(CaO)/w(SiO_2 = 3.5 \sim 5.5)$ for LF refining improves not only the desulfurization ability, but also the transformation of MgO-Al₂O₃ inclusions into Al₂O₃-MgO-CaO liquid inclusions.

Nowadays, the conventional method for modifying $MgO-Al_2O_3$ inclusions in industrial processes is to directly add calcium alloy to the molten steel. Whereas, the effectiveness of calcium treatment is limited due to the influence of the chemical properties of calcium and the composition of molten steel, and the castability of molten steel after calcium treatment is unpredictable. In addition, the calcium treatment leads to the form of new inclusions, increasing the amount of inclusions in steel and decreasing the cleanliness of molten steel. Moreover, the calcium treatment raises the cost of production, lowers the temperature of molten steel and lengthens the refining process. Meanwhile, the calcium treatment process emits a lot of dust, which is extremely unfriendly to the environment.

To address the issue of poor castability of LCAK steel produced by the "BOF-LF-CC" process and abolish the calcium treatment, a study aimed at improving the castability of LCAK steel was carried out, which mainly included laboratory studies focused on the interaction of refining slag and refractory materials with molten steel, as well as industrial trials to validate new processes. These investigations can provide the theoretical and experimental basis for improving the castability of the LCAK steel without calcium treatment.

2. Refining process of LCAK steel and evolutions of non-metallic inclusions

2.1. Refining of LCAK steel

The smelting process of LCAK steel is "BOF \rightarrow LF Refining \rightarrow CC", and the main operation and refining processes include: (1) During tapping of the converter, aluminum, ferromanganese alloy and lime were added for deoxidation, alloying and slag-making; (2) During LF refining, molten steel was heated in LF refining, and aluminum and lime were added for slagging, and the basicity of refining slag $w(CaO)/w(SiO_2)$ (C/S) was higher than 14, and the mass ratio of w(CaO) to $w(Al_2O_3)$ (C/A) was between 1.0 to 2.0; (3) Basicity of tundish flux was ≥ 2.0 and the mass ratio of w(CaO) to w(Al₂O₃) was about 1.2; (4) The ladle was clean without residue in the ladle. MgO-C refractory (w(MgO)>80%) were used in the slag line of the ladle, and the other part of the ladle adopted MgO-Al₂O₃-C bricks ($w(MgO)\approx 10\%$). The compositions of refining slag and molten steel used in the experiments are shown in TABLE 1 and TABLE 2, respectively.

TABLE	1
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Duo oogo	Content (wt.%)								C/S	
Frocess	SiO ₂	CaO	MgO	Al ₂ O ₃	MnO	P ₂ O ₅	T.Fe	S	C/8	C/A
LF refining beginning	6.10	45.64	4.89	38.03	1.24	0.06	3.61	0.27	14.72	1.09
LF refining ending	1.96	58.12	4.86	30.20	0.24	0.15	1.11	0.84	29.65	1.92
Casting in tundish	13.6	38.51	9.52	31.17	1.09	0.22	1.47	0.14	2.83	1.24

Chemical compositions of refining slag (wt.%)

TABLE 2

Compositions of LCAK steel (wt.%)

Process	C	Si	Mn	Р	S	Ca	Mg	Alt	Als
LF refining beginning	0.040	0.018	0.163	0.014	0.024	_	0.0004	0.040	0.032
LF refining ending	0.055	0.027	0.287	0.014	0.006	0.0008	0.0007	0.051	0.044
Casting in tundish	0.056	0.025	0.278	0.014	0.005	0.0006	0.0007	0.046	0.043

2.2. Evolution of non-metallic inclusions in LCAK steel

The samples of steel were taken at the stages of the LF refining beginning, the LF refining for 30 min, the LF refining ending, and the casting in tundish, respectively. Thereafter, the samples were detected by SEM-EDS. The results show that there are four types of oxide inclusions, that is, Al₂O₃, MgO-Al₂O₃, CaO-MgO-Al₂O₃ and CaO-Al₂O₃ inclusions, individually. The morphology of the inclusions and their proportions in LCAK steel are shown in Fig. 1 and Fig. 2, respectively.

It can be seen from Fig. 1(a) that the Al_2O_3 inclusions are mainly blocky or clustered, and the size of the inclusions is relatively large, ranging from 5 µm to 20 µm. The proportion of Al_2O_3 inclusions in the stage of LF refining beginning is relatively high (as shown in Fig. 2), being more than 90%. This is mainly due to the addition of a large amount of aluminum for deoxidation during the tapping. With continuous refining, the proportion of Al₂O₃ inclusions gradually falls to 33% after 30 min of LF refining. Meanwhile, the proportion of MgO-Al₂O₃ inclusions that leap to the main inclusions is close to 50%. The MgO-Al₂O₃ inclusions range in size from 2 μ m to 6 μ m and are mainly irregular, as shown in Fig. 1(b). However, at the stage of LF refining ending, the proportion of MgO-Al₂O₃ inclusions reduces, but it is still greater than 40%. As a result of the decrease of oxygen content in molten steel, [Al] in the molten steel reacts with MgO in the refractory or the refining slag to generate MgO-Al₂O₃ inclusions [4]. Meanwhile, the proportion of CaO-MgO-Al₂O₃ and CaO-Al₂O₃ in inclusions are 14% and 17%, separately, with sizes ranging from 2 µm to 6 µm. The CaO-MgO-Al₂O₃ inclusions have the same morphology as CaO-Al₂O₃ inclusions in that they are both spherical or quasi-



Fig. 1. Morphology of inclusions in molten steel; (a) Al₂O₃, (b) MgO-Al₂O₃, (c) CaO-MgO-Al₂O₃, (d) CaO-Al₂O₃



Fig. 2. Proportions of different inclusions

spherical. But, the morphology of inclusions $CaO-Al_2O_3$ is more regular, and the edges of $CaO-Al_2O_3$ inclusions are smoother, as seen in Figs. 1(c) and 1(d).

It can be seen from Fig. 2 that with the extension of refining time, the proportion of MgO-Al₂O₃ inclusions remains above 40% at the stage of the casting in tundish. This is mostly due to the tundish's refractory still contains MgO, and hence Al₂O₃ inclusions in the molten steel will continue to transform to MgO-Al₂O₃ inclusions [20]. The proportions of CaO-MgO-Al₂O₃ and CaO-Al₂O₃ inclusions increase from 2% to 15% and 3% to 20%, respectively. The reason for the increase of CaO-MgO-Al₂O₃ and CaO-Al₂O₃ inclusions, generally, is considered that a portion of CaO in the refining slag is reduced by [Al] in the molten steel to [Ca], and the reaction is shown as Eq. (4). Further, [Ca] reacts with Al₂O₃, MgO-Al₂O₃, and MgO-Al₂O₃-CaO inclusions, as shown in Eqs. (5)~(8). Consequently, the content of CaO in inclusions increase, and the content of MgO or Al₂O₃ in inclusions reduce [6,8,17,21].

$$2[Al] + 3(CaO) = 3[Ca] + (Al_2O_3)$$
(4)

$$[Ca] + n/3(Al_2O_3)_{\text{inclusion}} = = (CaO \cdot (n-1)/3Al_2O_3)_{\text{inclusion}} + 2/3[Al]$$
(5)

$$x[Ca] + (yMgO \cdot zAl_2O_3)_{inclusion} =$$

= (xCaO \cdot (y - x)MgO \cdot zAl_2O_3)_{inclusion} + x[Mg] (6)

$$[Ca] + (xCaO \cdot yMgO - zAl_2O_3)_{inclusion} =$$

= ((x + 1)CaO \cdot (y - 1)MgO \cdot zAl_2O_3)_{inclusion} + [Mg] (7)

$$[Ca] + (xCaO \cdot yMgO - z/3Al_2O_3)_{inclusion} =$$

= ((x + 1)CaO \cdot yMgO \cdot (z - 1)/3Al_2O_3)_{inclusion} +
+ 2/3[Al] (8)

Fig. 3 shows the evolution of CaO-MgO-Al₂O₃ inclusions in the LCAK steel by ternary phase diagram during LF refining. The region where the melting point of inclusions is lower than 1600°C is marked by a red curve. Fig. 3 indicates that the inclusions are mainly Al₂O₃ generated by deoxidation at the stage of LF refining beginning (Fig. 3(a)). Al₂O₃ inclusions transform into MgO-Al₂O₃ by Eqs. (1)~(3) during LF refining (Fig. 3(b)). After LF refining (Fig. 3(c)), even at the stage of the casting in tundish (Fig. 3(d)), the inclusions are mainly MgO-Al₂O₃ and a small amount of CaO-Al₂O₃ or MgO-Al₂O₃-CaO. MgO-Al₂O₃ inclusions in molten steel without calcium addition can easily adhere and accumulate at the head of the stopper during casting, which harms casting and even causes the interruption of casting [22].

Some enterprises installed ceramic filters in tundish to reduce the influence of $MgO-Al_2O_3$ inclusions on the castability of molten steel, however, the blockage problem of ceramic filters has not been adequately overcome after long-term usage [23]. Moreover, other materials for the immersion nozzle were also tested, but the results were not ideal [24].

3. The key factors for the formation of MgO-Al₂O₃ inclusions

According to Eqs. (1)~(3), the reason for the formation of MgO-Al₂O₃ inclusions is that MgO in the refractory or refining slag is reduced by [Al] to [Mg], and the generated [Mg] further reacts with [O] or Al₂O₃ inclusions in the molten steel. To improve the castability of molten steel, further investigations were carried out to quantitatively figure out the influence of refractories and refining slag on the formation of MgO-Al₂O₃ inclusions.



Fig. 3. Evolution of CaO-MgO-Al₂O₃ inclusions during LF refining; (a) LF refining beginning, (b) LF refining for 30 min, (c) LF refining ending, (d) Casting in tundish

The experiment was carried out in a vacuum induction furnace. Under the Ar atmosphere, 500 g LCAK steel was put into Al₂O₃-based (w(Al₂O₃) >99%) and MgO-based (w(MgO) >97.5%) crucibles, respectively. Then, the temperature of the furnace was raised to 1600°C. Aluminum and 50 g fluxes with different basicities were added to the molten steel to deoxidize and refine for 25 min. The molten steel was rapidly cooled in a vacuum furnace. The flux added in the experiments was made up of 25% Al₂O₃, 5% MgO, and the remaining CaO and SiO₂. The above chemical reagents were analytical pure (>99.99%) [25].

Three sets of group experiments were conducted in MgObased crucibles: one with no refining slag, and two with refining slag basicity of 2.5 and 3.5. Fig. 4 shows the composition of the inclusions in molten steel refined in MgO-based crucibles by Al_2O_3 -MgO-CaO ternary phase diagram. It can be seen from Fig. 4(b) and (c) that there are many MgO-Al_2O_3, as well as Al_2O_3 -MgO-CaO containing a small amount of CaO. In particular, although no refining slag (Fig. 4(a)) was added to the molten steel, MgO-Al_2O_3 inclusions were detected after 25 min of refining due to the use of MgO-based crucibles.

Besides, three sets of group experiments were carried out in Al₂O₃-based crucibles: one with no refining slag, and two with refining slag basicity of 4.5 and 8.5. Fig. 5(a) and (b) show that the inclusions in the molten steel with slag basicity less than 4.5 are primarily CaO-Al₂O₃ inclusions despite the slag containing 5% MgO. Meanwhile, as depicted in Fig. 5(a), a portion of inclusions formed without refining slag shows some CaO in their composition because the Al₂O₃-based crucibles are not very pure and may contain the impurity element Ca. As seen in Fig. 5(c), the addition of ultra-high basicity refining slag causes

the activity of MgO in the slag to increase, which in turn causes the formation of MgO-Al₂O₃ inclusions or Al₂O₃-MgO-CaO (with w(CaO) <5%) in molten steel.

It can be concluded from the above results that the transformation of Al_2O_3 to MgO- Al_2O_3 in the refining process has an important relationship with the composition of the ladle refractory and the basicity of refining slag. Therefore, the use of Al_2O_3 -based refractories and low basicity refining slag can effectively inhibit the transformation of Al_2O_3 to MgO- Al_2O_3 .

4. Industrial trials of improving the castability of LCAK steel

To improve the castability of LCAK steel, the production process of LCAK steel and ladle refractory has been optimized.

4.1. Composition optimization of ladle refractory

To reduce the influence of ladle refractory composition on the formation of inclusions in molten steel, the refractory of the ladle was replaced by MgO-Al₂O₃ with high Al₂O₃ content in this study. The refractory composition before and after optimization is shown in TABLE 3.

As shown in TABLE 3, the content of MgO in the ladle refractory is reduced significantly from 10.06% to 0.96%, reduced by more than 90%. In addition, considering the corrosion of refining slag on the ladle refractory, MgO-C refractory (w(MgO) > 75%) is still used in the slag line of the ladle [26].



Fig. 4. Composition distribution of the inclusions in CaO-MgO-Al₂O₃ system reacting using MgO-based crucibles; (a) Without slag, (b) R:2.5, (c) R:3.5



Fig. 5. Composition distribution of the inclusions in CaO-MgO-Al₂O₃ system using Al₂O₃-based crucibles; (a) Without slag, (b) R:4.5, (c) R:8.5

Comparison of ladle refractory composition before and after optimization

Type of refractory	SiO ₂	CaO	MgO	Al ₂ O ₃	T.Fe	С
Before optimization	2.00	1.29	14.06	82.03	0.15	0.25
After optimization	4.51	0.68	0.96	91.26	0.73	1.18

4.2. Production process optimization of LCAK steel

To reduce the load of desulphurization during LF refining, the process of "hot metal predesulphurization-BOF-LF-CC" was adopted in the industrial trails. The adoption of predesulphurization process reduces the sulphur content of molten steel in the converter from 0.022%~0.027% to 0.005%~0.010%, with an average reduction of 0.015%. The reduction of the sulphur content in the molten steel decreases the requirement of refining slag on the basicity, and the basicity of the refining slag was controlled to be between 3.5 and 5.5. Other operations are the same as the conventional process.

4.3. Industrial trials to improve the castability of LCAK steel

After the above optimization, the formation possibility of MgO-Al₂O₃ inclusions in the tundish still exists because the slag line portion of the ladle and the tundish still use MgO-containing refractories (the MgO content in the tundish refractories is about 60%). During the trials, to ensure the feasibility of casting, a "3 + 1" smelting pattern was adopted, that is, the molten steel is cast directly without calcium treatment in the first three heats, treated with calcium in the next heat, and the process was repeated.

The ladle with Al_2O_3 -based refractory was used for the production of LCAK steel. Fig. 6 shows the fluctuation of the molten steel level and the position of the tundish stopper during casting. After three furnaces of continuous casting, the position of the tundish stopper is increased by 10 mm without calcium addition. However, during the casting of the fourth furnace with calcium treatment, the position of the tundish stopper is reduced by 12 mm. It indicates the adherent substances consisting of MgO-Al₂O₃ and Al₂O₃-MgO-CaO to the stopper are dissolved by calcium-treated molten steel. However, the quality test results of the final steel product show that there is no significant difference between the cleanliness of the steel smelted in the fourth heat and that of the steel smelted by the conventional calcium treatment process. By the "3 + 1" smelting pattern, the number of continuous casting furnaces reaches 15 heats.

5. Conclusions

- Under the calcium-free treatment process, the evolution route of inclusions in the molten steel is Al₂O₃ → MgO-Al₂O₃ → CaO-MgO-Al₂O₃ → CaO-Al₂O₃ due to CaO in the refining slag with high basicity, but the transformation is not complete. The proportion of MgO-Al₂O₃ inclusions, which is the main factor that deteriorates the castability of molten steel, is still more than 40% after LF refining.
- (2) The inclusions in the molten steel are mainly MgO-Al₂O₃ inclusions after reaction for 25 min in a MgO-based crucible, even without refining slag. But the inclusions of the molten steel in the Al₂O₃-based crucible are mainly inclusions CaO-Al₂O₃, even if the top slag (basicity less than 4.5) contains about 5% MgO.
- (3) To reduce the proportion of inclusions MgO-Al₂O₃, the composition of ladle refractory and the basicity of refining slag are optimized, and the "3 + 1" process mode of LCAK steel is realized, that is, the molten steel is cast directly without calcium treatment in the first three furnaces, treated with calcium in the next furnace, and the process is repeated.



Fig. 6. Stopper position during continuous casting of LCAK steel

TABLE 3

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Conflict of interest All authors of this article have no conflict of interest.

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