Formation of Acicular Ferrite in Mg Treated Ti-bearing C–Mn Steel

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The effects of Mg content, inclusion size and austenite grain size on the acicular ferrite nucleation in Mg treated Ti-containing C–Mn steel were studied by high temperature experiment and metallurgical analysis. The composition analysis and size distribution counting of the inclusions and nucleuses were carried out on scanning electron microscope (SEM) with energy dispersive spectrometer (EDS) and image process software, separately. The effectiveness of inclusions with different size and the influence of heat treatment on acicular ferrite formation were statistically investigated. The results obtained are as fellows: the optimal Mg content for acicular ferrite nucleation is 0.0015–0.0026 mass%. The best austenite grain size for acicular ferrite nucleation is about 120 μm. The inclusions for nucleating acicular ferrite mainly consist of spherical Ti–Mg oxide covered by MnS, and there is a Mn-depleted zone (MDZ) around them. The optimized size of inclusions for acicular ferrite nucleation is between 1–4 μm in this study. The effectiveness of inclusions inducing acicular ferrite would be increased after heat treatment.

KEY WORDS: acicular ferrite; inclusion; nucleation; austenite grain size; size distribution.

1. Introduction

Grain refinement is the only method to enhance strength without deteriorating toughness in steelmaking. Since 1990 s, the Japanese metallurgical scholars proposed the technical theory of "Oxides Metallurgy" based on the function of inclusions in weld,[1] a lot of interests have been drawn to study the effect of inclusions during microstructure transformation process in steel. The acicular ferrite (AF), which nucleated intra-granularly on the surface of inclusions, has a chaotic crystallographic orientation.[2] This structure retards of the propagation of cleavage crack in the metal. In addition, it makes high strength steel have great toughness and good weldability.

A large number of studies indicated that the inclusions, such as Ti/Al/Zr oxides and Ti/Nb/V carbonitrides, could contribute to AF nucleation. So far, oxides metallurgy technology has already become an effective way to improve the performance of thick plate steel, low carbon non- tempered steel, high-strength low-alloy (HSLA) steel and welded joints of large heat input welding.[3–5] However, with the development of welding technology, welding heat input increasing causes the peak temperature rising up to 1 673 K, which makes the heat affect zone (HAZ) temperature too high and residence time too long, lots of TiN particles will redissolve into the γ-Fe matrix. Due to the lack of sufficient inclusion pinning grain boundary during period of high temperature, the austenite grain grows up rapidly and the welding performance of steel decreases remarkably.[6,7] So Ti oxides metallurgy cannot meet engineering and technology requirement.

Recently, growing concerns have been focused on trace Mg added into the steel.[8–11] Chang C. H. *et al.* and Kim H. S. *et al.* have studied the variation of inclusions and microstructures treated by Mg in Mn–Si–Ti deoxidized steel.[9,10] The cast microstructures of the steel, which contained large numbers of acicular ferrite, were obviously refined. Then, Wen B. *et al.* advocates that trace Mg was good for the heterogeneous nucleation of acicular ferrite during solidification.[11] Through the thermal simulation experiment, Chai F. *et al.* have studied the influences of Mg on the microstructures and impact toughness of the coarse grain heat affected zone (CGHAZ) in the Ti treated steel, they derived that about 0.002 mass% Mg added into Ti-containing steel was able to refine the Ti-based inclusions, which resulted in high volume percent of AF in CGHAZ.[12]

For now, there are few papers studying on the characteristics of nucleation inclusions for AF formation in Mg treated steel, especially on the best inclusion size for AF nucleation, and the influence of heat treatment on inclusion effectiveness of inducing AF nucleation. At present, the composition and size distribution of inclusions as well as the microstructure of ingot samples handled by trace Mg at 1 873 K have been investigated, and the optimal austenite grain size for AF nucleation and the variation of effectiveness of inclusions were explored.

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2. Experimental Procedure

The ingots were obtained by melting in a high temperature Molybdenum resistance furnace equipped with a corundum coated PtRh30-PtRh60 thermocouple and an FP93 series automatic temperature controller, by which the temperature error could be controlled within ±2 K. About 500 g raw Ti-containing C–Mn steel was melted within the corundum crucible with size of ϕ40 mm × 100 mm. After all materials melt, thermal insulation at 1 873 K for 5 min, treatment agents of Si–Mg alloy (contain 30 mass% Mg) were added into the melt, keeping the temperature for 5 min again, then turning off the power. When the temperature dropped to 1 473 K, the crucible was quickly taken out of the furnace and quenched into water. During the heating process, pure argon was pumped into the furnace in a flux of 1 L/min through the bottom to prevent oxidation, **Fig. 1** is the schematic diagram of experimental procedure. **Table 1** shows the chemical composition of prepared ingots. The Si, Mn, P, Al, Ti and Mg contents were measured by the method of ICP-AES; the C, S, N and total oxygen contents were obtained by middle infrared spectroscopy.

The samples, which size of ϕ8 mm × 10 mm, were sectioned from the middle of the ingots. They were mechanically ground, polished and then etched for 10–14 s using 4 vol% Nital.

Heat treatment tests were carried out in a box type high temperature silicon molybdenum resistance furnace. Two series of cube samples, size of 15 mm × 15 mm × 15 mm, were both heated to 1 173 K, 1 273 K, 1 373 K, and 1 473 K respectively, and hold for 20 min. One series of samples were air-cooled to room temperature to investigate the changes in microstructures and the other series of samples were water quenched to observe prior austenite grain size. All samples were ground and polished. The air-cooled samples etched for 10–14 s by 4 vol% Nital to display the microstructure, while the water quenched samples were etched at 353 K using a mixed agent (saturated picric acid + detergent + hydrochloric acid) to reveal the prior austenite grain boundary. The grain boundary and the microstructure of samples were observed by an optical microscope (OM). Inclusions in the samples were examined by SEM with an EDS.

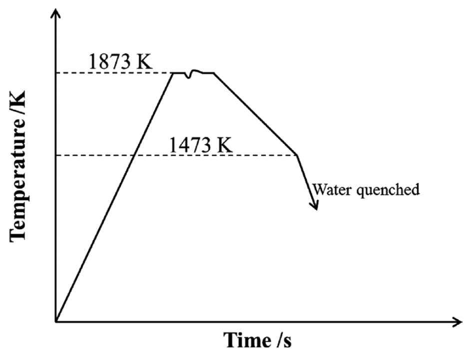

Fig. 1. The experimental procedure schematic diagram.

3. Results and Discussions

3.1. Effect of Mg Content on Microstructures

Figure 2 shows the microstructures of samples quenched from 1 473 K observed by OM. It could be found that there is a large number of ferrite side-plate (FSP) and a small amount of acicular ferrite (AF) in sample 1# as shown in Fig. 2(a), in which the content of Mg is 0.0005 mass%. When the content of Mg increase to 0.0015 mass% and 0.0026 mass%, the dominated microstructures of samples 2# and 3# changed into acicular ferrite (Figs. 2(b) and 2(c)). As the Mg content up to 0.0054 mass% in sample 4#, the microstructure turned into FSP again, the amount of acicular ferrite is appreciably reduced (Fig. 2(d)). From the foregoing that a certain content of Mg can promote acicular ferrite formation in Mg treated Ti-containing C–Mn steel. The optimized Mg contents for acicular ferrite nucleation is found to be 0.0015–0.0026 mass% by current experiment. As can be seen from **Fig. 3** that when the Mg content increasing, the inclusion number percentage, size of <3 μm, first increase then decrease, the inclusion mean size first decrease then increase. This is not exactly in keeping with the result of H. S. Kim *et al.*,[10] and the difference may come from the long holding time after Mg added into the melt in present study, during which Mg-containing inclusions are susceptible to growth by collision and agglomeration. The number percent of inclusion with size of <3 μm in sample 2# and 3# is the highest, which means the inclusion for AF nucleation is more, so the AF content in two samples is higher.

3.2. Effect of Inclusion Size on the AF Nucleation

The size distribution of inclusions has an important effect on acicular ferrite nucleation. There are a lot of reports announced about the effect of inclusion size on heterogeneous nucleation of acicular ferrite in recent years. Barbaro F. J. *et al.* stated that acicular ferrite could formed easily when the diameter of inclusions were in the range of 0.4–0.6 μm.[13] Yamamoto K. *et al.* demonstrated that the inclusion size of 0.4–2.0 μm was effective to acicular ferrite nucleation.[14] Lee T. K. found that the inclusions with diameters of 0.25–0.8 μm have a good nucleation potential.[15] His calculations found that the inclusion nucleation probability increases with the size of inclusions when the inclusion size

Table 1. The chemical compositions of ingots in mass%.

	C	Si	Mn	P	S	Ti	Mg	Al	N	T.O
1#	0.13	0.28	0.99	0.025	0.027	0.0090	0.0005	<0.005	0.0023	0.0041
2#	0.13	0.29	0.99	0.025	0.027	0.0073	0.0015	<0.005	0.0019	0.0034
3#	0.13	0.35	0.99	0.025	0.027	0.0078	0.0026	<0.005	0.0021	0.0023
4#	0.13	0.38	0.99	0.025	0.027	0.0087	0.0054	<0.005	0.0025	0.0014

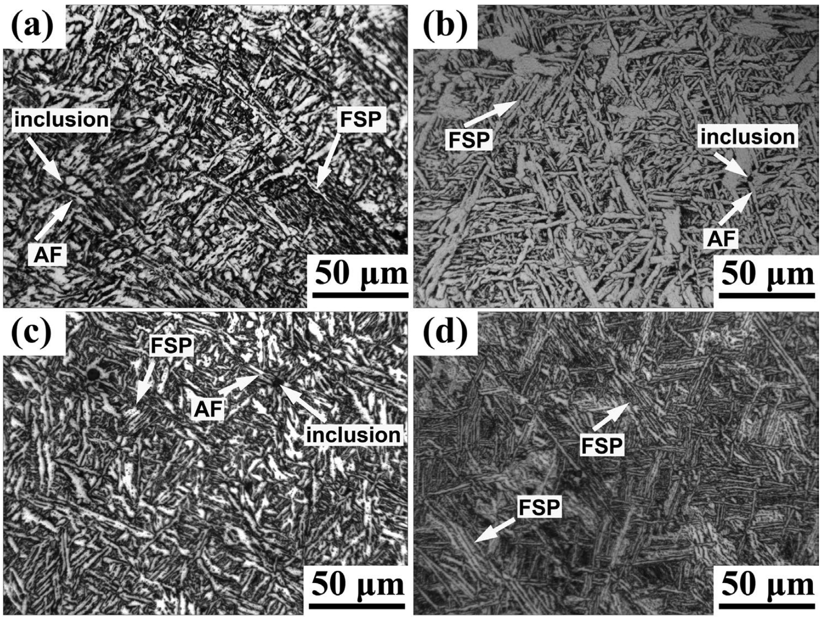

Fig. 2. Microstructures of steels quenched from 1 473 K with different Mg content. ((a) [Mg] = 0.0005%, (b) [Mg] = 0.0015%, (c) [Mg] = 0.0026%, (d) [Mg] = 0.0054%).

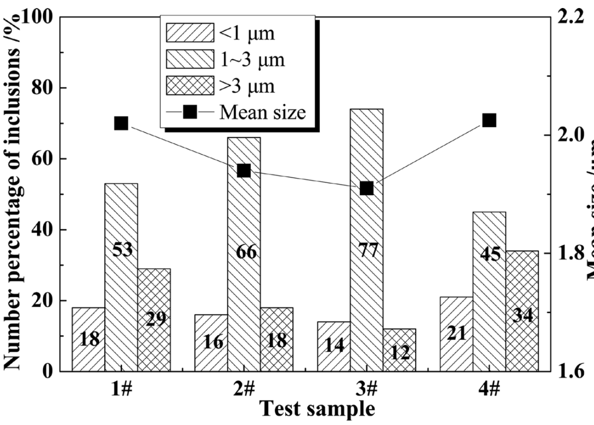

Fig. 3. The inclusion size distribution of four test steels.

smaller than 1.1 μm.

Figure 4 shows the situation of AF nucleation and the inclusions in sample 3# under the state of ingot and heat treatment. It can be seen that a lots of inclusions have the function of promoting acicular ferrite nucleation in both specimens. Acicular ferrite content in the sample heat treated at 1 473 K was higher than that in the ingot. Most of the fine inclusions have the ability of inducing acicular ferrite, but a few bigger ones have the same effect. The AF laths induced by small inclusions are obviously smaller than the AF laths induced by the big inclusions. It means that the size of the nucleation inclusion has great influence on the size of AF lath.

In order to observe the size distribution of the inclusions, taking 30 photos randomly at different fields of 500 times magnification by scanning electron microscope from sample 3#. All photos were analyzed by the image software statistically. **Figure 5**(a) exhibits the inclusions size distribution of sample 3# of the cast and heat treated samples, separately. As seen in the figure, the amount percentage of the inclusions with the size of <1 μm, 1–2 μm, 2–3 μm, 3–4 μm and >4 μm was 17.3%, 31.5%, 39.1%, 8.1% and 4.1% in the cast of sample 3#, respectively. The quantity percentage of the inclusions size of 3–4 μm and >4 μm changed into 10.8% and 12.7% in the heat treated sample. The size of inclusions >3 μm in the ingot grow after heat treatment process, simultaneously the amount of inclusions size of 2–3 μm decreased to 27.2%. It deserves to be mentioned that the amount change of inclusions with size of <1 μm and 1–2 μm were not significant in both sections, they were 18.0% and 31.4% after heat treatment, respectively. It can be found that the inclusion size in Mg treated C–Mn–Ti steel is very small, mainly concentrate in 1–3 μm.

More than 200 nucleuses in the ingot sample and 300 nucleuses in the heat treated sample were counted. Image process software was used to analyze the size distribution of the cores in both samples. As shown in Fig. 5(b), among the inclusions inducing acicular ferrite nucleation in cast sample 3#, the inclusions with the size of <1 μm accounts for 8.0%, while inclusions size in 1–2 μm and 2–3 μm were 35.2% and 46.2%, respectively. While the quantity of nucleant inclusions size of 3–4 μm decreased to 7.5% obviously. For the inclusions bigger than 4 μm the corresponding proportions was 3.1%, the amount was decreased sharply. After austenized at 1 473 K and cooled by air, the percentage of inclusions with the size of <1 μm, 1–2 μm, 2–3 μm, 3–4 μm and >4 μm were 8.3%, 35.7%, 37.2%, 10.2% and 8.6%, respectively. Therefore, the size of the inclusions inducing acicular ferrite nucleation is mainly concentrated in 1–3 μm in present steel. The quantity of big inclusions that can induce acicular ferrite is increased after hold at 1 473 K for 20 min.

The ratio of nucleant inclusions to total inclusions in each size range can exhibit the corresponding nucleation ability of inclusions in each size. **Figure 6** presents the amount ratio of nucleant inclusions to total inclusions in each size range of the ingot and heat treated sample 3#. It can be seen from that the nucleation ratio of inclusions size of <1 μm were only 28.9% and 30%, respectively. The nucleation ability increased when inclusion size was 1–2 μm, the

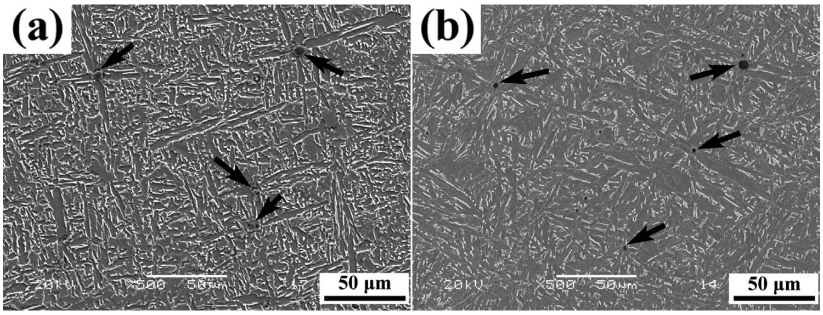

Fig. 4. The situation of AF nucleation on inclusions of sample 3#. (a) as-cast and (b) heat treated at 1 473 K.

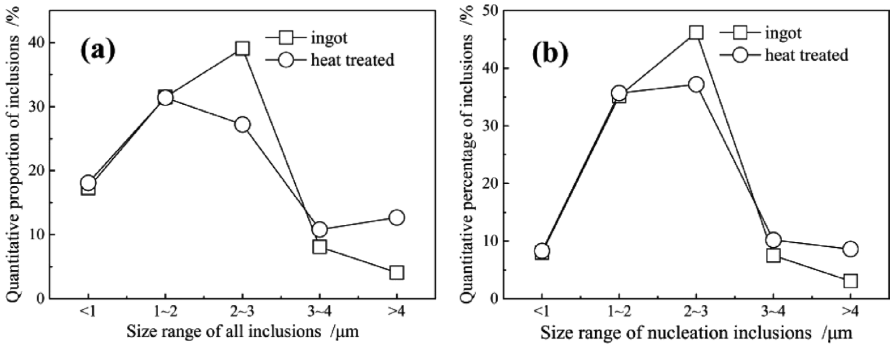

Fig. 5. The size distribution of nucleant inclusions and all inclusions in the ingot and heat treated of sample 3#. (a) all inclusions and (b) nucleant inclusions.

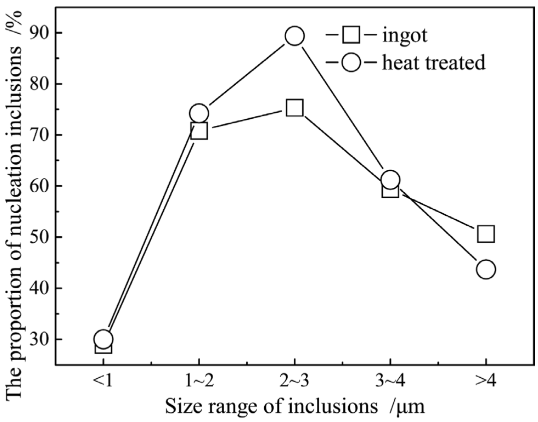

Fig. 6. The ratio of nucleation inclusions in each size range.

proportion of nucleation inclusions were 70.8% and 74.2%, respectively. And the inclusions size of 2–3 μm had the largest percentage of nucleant inclusions were 75.3% and 89.4%, respectively. The nucleation ability of the inclusions after heat treatment was obviously improved, compared to the ingot. This is consistent with the previously discussed results of microstructure in Fig. 4. Although the nucleation ratio of both section were decreased to 59.4% and 61.2% when inclusion size further increased to 3–4 μm, the nucleation ability of inclusions in the heat treated sample was still better than the cast. It was worth noting that the nucleation ratio for inclusions size of >4 μm only accounted for 50.6% and 43.7%, after heat treated the nucleation ability of inclusions decreased obviously, it was lower than the inclusions of ingot in this range. It could be concluded that after heat treated at 1 473 K for 20 min, the nucleation ability of inclusions in sample 3# was improved when the inclusion size was 1–4 μm. In order to increase the number of nucleation inclusions in the actual control of Mg treatment process, it is important to control the size of inclusions in the range of 1–4 μm.

3.3. Effect of Grain Size on the AF Nucleation

It could be clearly seen that the sample 3# is the best choice to study the law of acicular ferrite nucleation in Mg treated steel. The ingots of sample 3# were cut into many sections with the size of ϕ10 mm $\times$10 mm for the heat treatment experiments. They were mechanically ground and polished for metallographic observation and analysis.

Figure 7 shows the air-cooled microstructures of the sample 3# austenitized at different temperatures for 20 min. It can be seen that the microstructure austenitized at 1 173 K of sample 3# consists of a lot of block ferrite and pearlite. When the austenitizing temperature rose up to 1 273 K, the amount of block ferrite and pearlite decreased, and some acicular ferrite generated in the gap of the block ferrites. When the austenitizing temperature was 1 373 K, acicular ferrite content further increased and the proportion of block ferrite and pearlite appreciably decreased. As the austenitizing temperature increased to 1 473 K, the microstructure contains lots of acicular ferrite and a little block ferrite. Therefore, it can be concluded that the best austenitizing temperature is 1 473 K at present study. After austenized 20 min at 1 473 K and then cooled by air, the Mg treated Ti-bearing C–Mn steel can obtain a large amount of acicular ferrite.

The acicular ferrite nucleation is related with not only the composition of inclusions but also many other factors, such as austenite grain size, size distribution of inclusions and the cooling rate from 1 073 K to 773 K. It is reported that when the austenite grain size reaches a certain value, the volume-maximizing of acicular ferrite could be realized.[17] There were many studies indicated that for different composition of steel, the optimum austenite grain size in favor of acicular

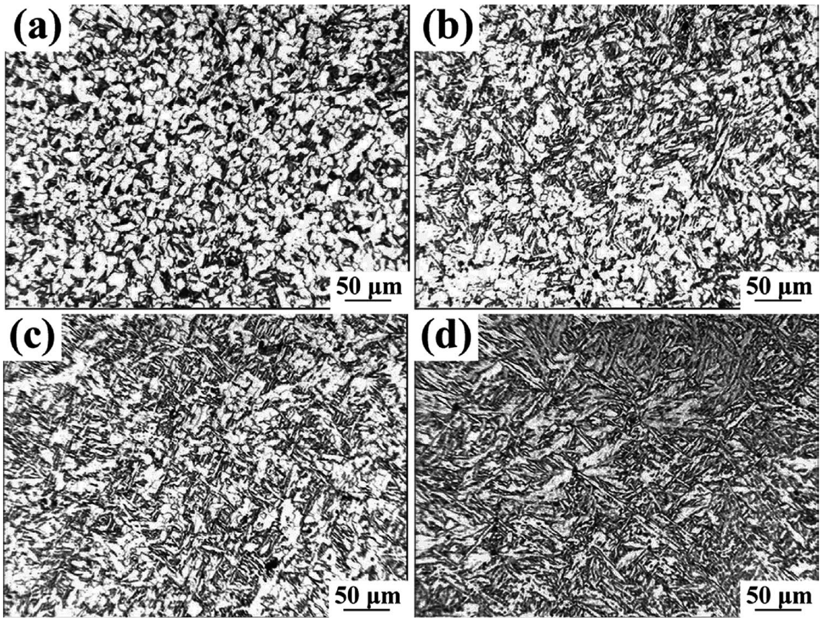

Fig. 7. The microstructures of sample No. 3# hold for 20 min at different austenitizing temperatures then cooled by air. (a) T=1 173 K, (b) T=1 273 K, (c) T=1 373 K, (d) T=1 473 K.

ferrite nucleation is different. Barbaro F. J. *et al.*[13] maintained that the optimal austenite grain size was around 100 μm which could benefit the acicular ferrite formation. Lee J. L. *et al.*[16] found that when the austenite grain size is larger than 100 μm, it contributes to acicular ferrite nucleation. So it is necessary to consider the optimum austenite grain size for acicular ferrite nucleation in Mg treated Ti-containing steel. Under the same soaking time, various austenitizing temperature, the corresponding austenite grain size would not be the same. The content of acicular ferrite in sample 3# was the highest when the austenite temperature is 1 473 K. It means that the austenite grain size is the optimum to nucleate acicular ferrite. **Figure 8** shows the primary austenite grain boundary of sample 3# after holding for 20 min at the austenizing temperature of 1 473 K, in which the primary austenite grain size is about 120 μm. Therefore, for the steel treated by Mg, the best austenite grain size which promotes the formation of acicular ferrites is approximately 120 μm.

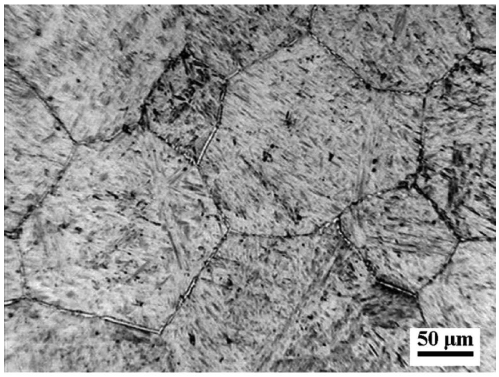

Fig. 8. The austenite grain of sample 3# hold for 20 min at austenitizing temperature of 1 473 K.

3.4. Mechanisms of AF Nucleation

So far, a number of mechanisms have been proposed to explain the function of non-metallic inclusions on the nucleation of acicular ferrites: (1) a reduction in the interfacial energy for simple heterogeneous nucleation on the surface of inclusion;[17] (2) an epitaxial nucleation on the inclusion external surface, inclusion lattice have a good coherency with ferrite;[18] (3) increasing stress around the inclusions resulted from the differences in thermal expansion coefficients of the inclusions and matrix metal;[19] (4) solute depletion of elements in the matrix near inclusions, such as Mn, C.[20,21] The inert interface mechanism is not considered at present, as it is quite difficult to looked into yet and few studies have conducted. The average thermal expansion coefficient of Mg-containing inclusions are 10^{-5}–10^{-6} /K (*e.g.* MgO, 13.5×10^{-6} /K) in the range of 273–1 273 K. The thermal stress between steel matrix and inclusion is, about 10^8 J/mol, insufficient to induce AF nucleation. The lattice misfits between Mg-containing inclusions and α-Fe are very small, such as $MgTi_2O_4$ (1.47%), Mg_2TiO_4 (1.84%), MgO (4.03%), which may plays a stronger role in AF nucleation. For Mn-depleted zone (MDZ) mechanism of acicular ferrite nucleation, it is well known that the precipitation of MnS on the surface of Ti–Mg complex oxides inclusion would decrease Mn concentration in the matrix metal near the inclusion. It is well established that Mn content increasing would enlarge the austenite zone. Therefore, Mn concentration decreasing in the matrix near the precipitated MnS inclusions would reduce the austenite zone and contributes to the nucleation of ferrite.

The morphology and composition of the inclusion inducing acicular ferrite nucleation in sample 3# is shown in **Fig. 9**. It can be found that a typical acicular ferrite lath nucleated on the inclusion surface, and the ferrite lath can induce more ferrite formation in different direction due to the sympathetic nucleation mechanism. The morphology of the inclusion is nearly spherical complex particle. From the spectra patterns of EDS, it can be known that the inclusion is composed by Ti/Mg oxide inside, and MnS covered outside. **Figure 10** is the X-ray line scanning microanalysis result of the complex particle and matrix metal, it can be found that there is a Mn-depleted zone patently around the

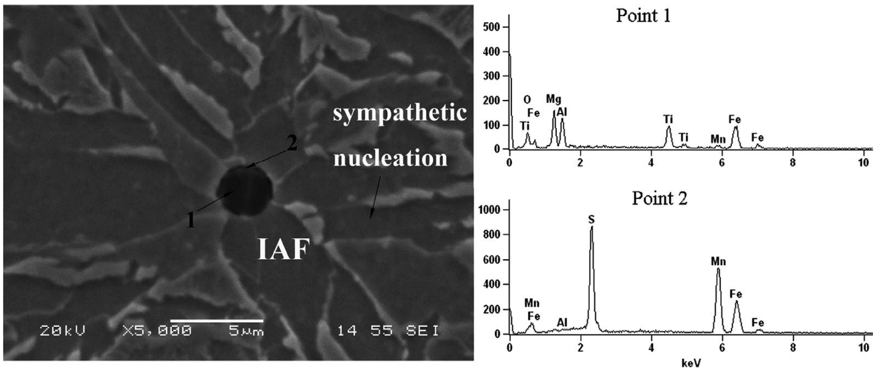

Fig. 9. The morphology and composition of the acicular ferrite nucleation inclusion.

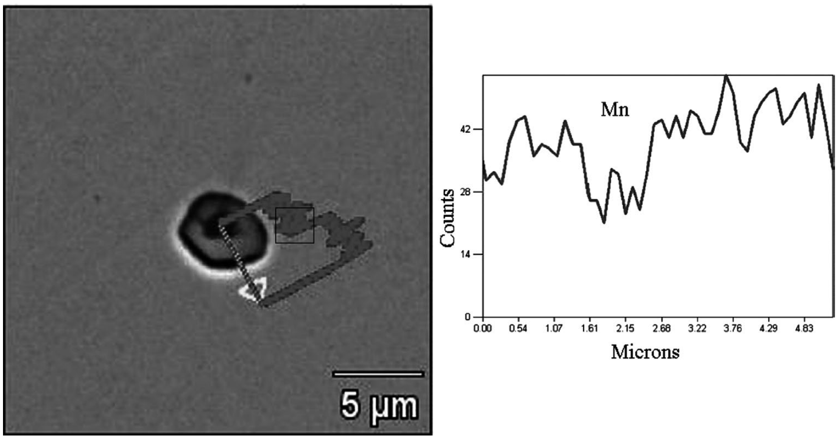

Fig. 10. The line analysis of Mn in complex inclusion and matrix.

complex inclusion. So the combined effects of MDZ and small misfit maybe the main mechanism of Mg-containing inclusion inducing AF nucleation effectively.

4. Conclusions

The effect of the Mg content on microstructure, and the influence of heat treatment on nucleation effectiveness of inclusion were investigated using Mg treated Ti-bearing C–Mn steel. The results obtained are as follows:

(1) Inclusions in Mg treated C–Mn–Ti steel are effective on acicular ferrite nucleation. The optimum Mg content is 0.0015–0.0026 mass%. The inclusions inducing acicular ferrite nucleation is composite inclusions consisting of Ti–Mg oxide core and MnS shell, and around the complex inclusion there is a Mn-depleted zone. Mn-depleted zone and small misfit can be considered as the main mechanism of complex inclusion inducing acicular ferrite.

(2) A large amount of acicular ferrite is achieved in the Mg teated C–Mn–Ti steel. The best austenizing temperature is 1 473 K. The optimal austenite grain size is about 120 μm which is good for AF nucleation.

(3) The favorable inclusion size for acicular ferrite nucleation is in range of 1–4 μm for the Mg teated C–Mn–Ti steel. The heat treatment process can improve the nucleation ability of inclusions in this range obviously.

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