A study on the acicular ferrite formation in steel weld metals for gas metal arc welding*

by Kyohei Uto**, Koyo Nakayama**, Yuji Kisaka***, Fumiaki Kimura***, Hidenori Terasaki****

Characteristics of oxide inclusions in steel weld metals with varying acicular ferrite (AF) fractions, which were produced by gas metal arc welding using controlled CO2 (10%, 30%, and 50%) and titanium contents (by placing ultra-fine Ti wire), were statistically investigated. The correlation between identified phases in the oxide inclusions and AF formation was discussed from the viewpoint of AF formation mechanism. For high AF fraction samples, we confirmed that all oxide inclusions include Mn-Si-Al-Ti-O amorphous phases. By contrast, an amorphous phase was never observed for low AF fraction samples. Additionally, we confirmed that a few of the amorphous oxides created a Mn-depleted zone (MDZ), which suggested that the MDZ formed by Mn-Si-Al-Ti-O amorphous phase stimulates AF formation.

Key Words: acicular ferrite, gas metal arc welding, inclusion, amorphous, manganese depleted zone, spinel oxides

1. Introduction

Acicular ferrite (AF) in low-carbon steel weld metals is regarded as the most desirable microstructure with respect to strength and toughness. Hence, research on the mechanism of AF formation is required to further improve the mechanical properties of steel weld metals. Inclusions in weld metal are known to contribute to the nucleation of AF.1,2) TiO,1,3) TiN,1) and spinel oxides 4,5) have often been noted as effective nucleants for AF. Generally, these inclusions are evaluated using lattice matching mechanism. These inclusions exhibit a low lattice misfit by satisfying the Baker–Nutting (B–N) orientation relationship with ferrite.3) It has been suggested that inclusions with a low lattice misfit located interface between inclusion and matrix play an important role in AF nucleation. However, some studies have reported that TiN with a low lattice misfit is not an AF nucleant.6) Additionally, Nako et al.7) reported that it is difficult for the Kurdjumov–Sachs (K–S) relationship, which describes the orientation between ferrite and austenite, and the B–N orientation relationship to coexist.

Ti2O3 and MnS are also reported as effective inclusions for the formation of AF; however, these inclusions do not have a low lattice misfit with ferrite. Instead, these materials are believed to promote AF formation by creating Mn-depleted zones (MDZs) around inclusions.2) MDZs are considered to arise from the penetration of Mn atoms into the atomic vacancies of Ti2O3 and from the formation of MnS in the case of MnS. MDZs formed around inclusions are believed to enhance the transformation driving force to ferrite and promote AF nucleation. However, contradictory findings have been reported, in which MDZs were not observed around Ti2O3 and MnTi2O4 which are generally believed to form MDZs.7, 8) Although Ti-containing oxides are expected to play an important role in AF formation, discrepancies and contradictory findings on their role in AF nucleation necessitate the need for further investigation. In this study, the quantity of AF formed and the characteristics of the produced oxide are adjusted by controlling the CO2 content in the shield gas for gas metal arc welding (GMAW) along with the level of Ti added to the weld metal. Further, we aim to elucidate the AF formation mechanism by analyzing its chemical composition and crystal structure. Based on our results, we propose the effect of Mn-Si-Al-Ti-O amorphous phase as a new mechanism for AF formation.

2. Experimental procedure

Samples were produced by multi-layer GMAW. API 5L X65 was used for the steel pipe and AWS A5.18 ER70S-G was used for welding wire(1.2φ). Four weld metals A, B, C, and D were investigated. Sample A had a shield gas composition of 70% Ar and 30% CO2; sample B had a shield gas composition of 50% Ar and 50% CO2; sample C had a shield gas composition of 90% Ar and 10% CO2. Sample D had the same shielding gas composition as sample A; however, the level of additional Ti was increased. This was carried out by placing additional ultra-fine Ti wire to the existing welding wire in the welding line. Table 1 shows the chemical composition and welding conditions of the weld metal determined for the first layer of the weld metal.
The microstructures of the weld metals were examined using a scanning electron microscope–electron backscatter diffraction (SEM-EBSD) method. An acceleration voltage of 15 kV and a step size of 0.2 μm were employed for the measurement. A closed area where the crystal orientation difference was less than 15° was defined as a grain. The grain size was defined as the average circle equivalent diameter of the observed area. The AF fraction was defined as the fraction of area for which the equivalent circle diameter of the grain is 0.1–10 μm. The AF grain size was defined as the average circle equivalent diameter in the area (50 μm * 50 μm) occupied by AF. The microstructures of the weld metals were observed from two fields of view for the first layer. Further, the AF fraction, grain size, and AF grain size values of each sample were given as the average of the two fields of view.

For each sample, the oxides on the first layer were collected as thin foils using a focused ion beam (FIB) device and then observed by transmission electron microscope (TEM). The chemical composition distribution was analyzed by scanning transmission electron microscope–energy-dispersive X-ray spectrometry (STEM-EDS).

3. Result and discussion

3.1 Microstructural evaluation

The microstructures were observed by SEM-EBSD. Figure 1 shows the grain maps and grain sizes in each area; Figures 1(a)–(d) show grain maps and grain size. Sample A exhibited the smallest grain size among the four samples. Although sample B also exhibited a small microstructure, the grain size of sample B was larger than that of sample A because of the coarse grain boundary ferrite. Overall, samples C and D displayed coarse grains and a large grain size. Figures 1(e)–(h) show grain maps and AF grain size. AF grain size is average circle equivalent diameter of the area shown in (e)-(h). Sample B displayed the smallest AF grain size among the four samples, while sample C had the largest AF grain size. Figure 2 shows the average AF fraction for each sample. The average AF fraction was high for samples A and B and low for samples C and D.

![Fig. 2 The average AF fraction for each sample](image)

3.2 Oxide observation for high AF fraction samples (samples A and B)

Seven and six oxides were extracted from samples A and B using FIB, respectively. Figure 3 shows the typical elemental analysis results from STEM-EDS for these oxides. These results

![Fig. 3 Oxide observation for high AF fraction samples](image)
demonstrated that Al-Ti-Mn oxides surrounded by Si oxides were present within the sample. Moreover, Mn and S phases were observed at the interface between inclusion and matrix in some oxides and identified as MnS by selected area diffraction pattern (SADP) analysis. Additionally, a Ti-rich layer was present at the interface between inclusion and matrix of some oxides.

For all 13 oxides, Al-Ti-Mn oxides and Si oxides were present inside oxide inclusions. Some oxides contained Al-Ti oxides instead of Al-Ti-Mn oxides and were commonly surrounded by Si oxides. However, the presence of a Ti-rich layer and MnS differed depending on the oxide. The Ti-rich layers were evaluated by line analysis because it has been suggested that the existence of Ti oxides exhibiting a low lattice misfit with ferrite is important for AF formation. Figure 4 shows the line analysis results of the oxide interface where a Ti peak was observed outside the Si oxides. Ti-rich layers were confirmed in eight oxides.

Next, the oxide phases were investigated by TEM. Figure 5 shows dark-field (DF) images and SADP results of the identified phases which corresponds to Al-Ti-Mn oxides observed in Fig. 3. The spinel oxides (MnAl2O4) were observed in samples A and B as shown Fig. 5. Figure 6(a) shows the bright-field(BF) image of the oxide shown Fig. 3(b), and Fig. 6(b) shows a lattice image of the Si oxides within the inclusion. This image shows that the lattice arrangement for the Si oxides determined by EDS was irregular. Further, we performed a fast Fourier transform (FFT) of the lattice image corresponding to the position of Si oxides as shown in Fig. 6(c). This figure shows an amorphous halo pattern. From these results, it can be concluded that the Si oxides were amorphous phases of Mn-Si-Al-Ti-O.

3.3 Oxide observation for low AF fraction samples (samples C and D)

Fig. 3 Typical elemental analysis results for oxide extracted from (a)sample A and (b)sample B

Fig. 4 Line analysis result for oxide with Ti-rich layer at interface between inclusion and matrix. The analysis position is shown orange line in (a) and the direction is from inclusion to matrix as shown red line in (a).

Fig. 5 (a)SADP of MnAl2O4, (b)DF image formed by light source inside yellow circle of (a)

Fig. 6 (a) BF image of oxide with Si oxides (b) lattice image in the red box of (a) (c) image after FFT in the red box of (b)
Six and seven oxides from samples C and D were extracted using FIB, respectively. Figure 7 shows typical elemental analysis results for these oxides; all 13 oxides exhibited similar patterns and were wholly occupied by Al-Ti oxides. Additionally, MnS and Ti-rich layers were confirmed at the interface between inclusion and matrix for some oxides.

In contrast to samples A and B, Si oxides (Mn-Si-Al-Ti-O amorphous phases) and Mn-containing oxides (excluding the MnS phase) were never observed in oxide inclusions. Figure 8 shows a DF image and SADP results for the identified phases. The corundum oxides (Ti₂O₃) were observed in sample D.

3.4 The effectiveness of the observed elements

Based on the microstructural results showed in Section 3.1, samples A, B, and C, D are considered as high AF fraction and low AF fraction samples, respectively. Figure 9 and Table 2 summarize the feature of the oxides in each sample; the effectivenesses of the oxides were evaluated with reference to these features.

First, the MnS phases were confirmed at the interface between inclusion and matrix in both high AF fraction and low AF fraction samples, with nearly identical percentage. This finding suggested that MnS is unlikely to contribute to AF nucleation in this study.

Second, there was a difference in the presence of Ti-rich layers, in that, they were more frequently observed high AF fraction samples compared to low AF fraction samples. However, amorphous occupied a lot at the interface between inclusion and matrix, and Ti-rich layer existed only partially. Hence, it seems unlikely that the Ti-rich layer directly affected AF nucleation.

Third, there was a difference in the presence of spinel oxides. The spinel oxides (MnAl₂O₄) were only observed in samples A and B. The corundum oxides (Ti₂O₃) in the place of spinel oxide were detected in sample D. This result was in agreement with the findings of Wang et al.,[7] who reported that spinel oxides were rarely observed and that pseudo-brookite and corundum oxides were generated in a sample containing a large quantity of Ti based on thermodynamic calculation results. However, in this study, the spinel oxides were present within the oxides rather than at the interface between inclusion and matrix, and they were surrounded by amorphous phase. Hence, it seems unlikely that the spinel oxides contribute to AF nucleation by lattice matching mechanism.

Finally, the biggest difference was based on the presence of amorphous phases. Mn-Si-Al-Ti-O amorphous phases were present in all oxides of high AF fraction samples but were not observed in the oxides for low AF fraction samples. The absence of Mn-Si-Al-Ti-O amorphous phases in samples C and D can be explained by the Ellingham diagram. In the equilibrium state, the free energy of formation of metal oxides was Al, Ti, Si, and Mn in the order of lower energy, i.e., O bonds with the metals in this order formed oxides. Because the oxygen content was lower for sample C compared with samples A and B, O bonds with the metals in this order formed oxides. Because the oxygen content was lower for sample C compared with samples A and B, O bonds with the metals in this order formed oxides. Because the oxygen content was lower for sample C compared with samples A and B, O bonds with the metals in this order formed oxides. Because the oxygen content was lower for sample C compared with samples A and B, O bonds with the metals in this order formed oxides. Because the oxygen content was lower for sample C compared with samples A and B, O bonds with the metals in this order formed oxides. Because the oxygen content was lower for sample C compared with samples A and B, O bonds with the metals in this order formed oxides. Because the oxygen content was lower for sample C compared with samples A and B, O bonds with the metals in this order formed oxides. Because the oxygen content was lower for sample C compared with samples A and B, O bonds with the metals in this order formed oxides.

Therefore, Mn-Si-Al-Ti-O amorphous phases were not formed in the oxides for samples C and D. From the results of this investigation, it is suggested that the presence of Mn-Si-Al-Ti-O amorphous phases are important for AF formation. Based on this result, we considered the mechanism of AF formation. Lattice matching theory is unlikely when an amorphous phase occupies the space around the oxides, because an amorphous phase has an irregular lattice arrangement and does not exhibit a low lattice misfit with ferrite. In contrast, some Mn-Si-Al-Ti-O amorphous phases formed MDZs between the amorphous phases and ferrite as shown.
4. Conclusion

The findings of this study can be summarized as follows:

(1) An evaluation of the AF microstructure demonstrated that samples with a large quantity of additional Ti and a decreased CO₂ percentage in the shielding gas reduced AF fraction. Moreover, the samples with an increased CO₂ percentage in the shielding gas exhibited finer AF grains.

(2) MnS was found in some oxides for all samples regardless of AF formation. Thus, MnS is unlikely to contribute to AF nucleation.

(3) Amorphous occupied a lot at the interface between inclusion and matrix, and Ti-rich layer existed only partially. Thus, the presence of a Ti-rich layer at the interface is unlikely to contribute to AF nucleation.

(4) Although spinel oxides were observed only in the high AF fraction samples, they were located inside the oxides. Thus, the spinel oxide is unlikely to contribute to AF nucleation.

(5) In the high AF fraction samples, Mn-Si-Al-Ti-O amorphous phases were observed at the interface between inclusion and matrix. As some Mn-Si-Al-Ti-O amorphous phases were at the interface between the amorphous phase and matrix with MDZ, it is suggested that MDZ formed by Mn-Si-Al-Ti-O amorphous phases is a preferable mechanism to stimulate AF formation.

Reference

1) Mills A. R., G. Thewlis, and J. A. Whiteman. "Nature of inclusions in steel weld metals and their influence on formation of acicular ferrite." Materials Science and Technology 3.12 (1987): 1051-1061.

2) DS Sarma, Karasev AV, and Jönsson PG. "On the role of non-metallic inclusions in the nucleation of acicular ferrite in steels." ISIJ International 49.7 (2009): 1063-1074.

3) Yamada Tomonori, Hidenori Terasaki, and Yu-ichi Komizo. "Relation between inclusion surface and acicular ferrite in low carbon low alloy steel." ISIJ International 49.7 (2009): 1059-1062.

4) Wang Bingxin, Xianghua Liu, and Guodong Wang. "Inclusion characteristics and acicular ferrite nucleation in Ti-containing weld metals of X80 pipeline steel." Metallurgical and Materials Transactions A 49.6 (2018): 2124-2138.

5) Kang Yongjoon, et al. "Factors affecting the inclusion potency for acicular ferrite nucleation in high-strength steel welds." Metallurgical and Materials Transactions A 47.6 (2016): 2842-2854.

6) Shim J-H., et al. "Ferrite nucleation potency of non-metallic inclusions in medium carbon steels." Acta Materialia 49.12 (2001): 2115-2122.

7) Nako Hidenori, et al. "Crystal orientation relationships between acicular ferrite, oxide, and the austenite matrix." ISIJ International 54.7 (2014): 1690-1696.

8) Kang Yongjoon, et al. "Mn-depleted zone formation in rapidly cooled high-strength low-alloy steel welds." Metallurgical and Materials Transactions A 45.11 (2014): 4753-4757.

9) Loder Denise, Susanne K. Michelie, and Christian Bernhard. "Acicular ferrite formation and its influencing factors—a review." J Mater Sci Res 6.1 (2017): 24-43.

10) Seo Kangmyung, et al. "Formation of Mn-depleted zone in Ti-containing weld metals." Welding in the World 59.3 (2015): 373-380.