Inclusion core in the weld pool of HSLA steel was directly observed by Time Resolved X-Ray Diffraction experiments. The aluminum concentration to oxide was changed from 0.48 to 1.52 through the experiments. The corundum alumina was identified in the weld pool and the population increased as the contents of aluminum increased. The results of TRXRD experiments was related to the direct observation of inclusion by TEM analysis and the formation process of inclusion in HSLA steel was summarized in the sense of overall microstructure evolution during welding. When an aluminum content was approaching stoichiometrical value to oxygen level, the corundum alumina reacted with glassy phase and spinel-type oxide was formed after solidification process of welding.

KEY WORDS: inclusion; HSLA steel; deoxidization; synchrotron radiation; welding.

1. Introduction

The acicular ferrite is ideal microstructure for the weld metal of high strength steel. The weld metal consisting of the microstructures possesses the high tensile strength (~700 MPa) and toughness even if the alloying level is low. The superiority of mechanical properties is due to small grain size and the multi-directional distribution of ferrite plates. It is well-known that the nucleation site of acicular ferrite is inclusion in the weld metal.1) As the inclusion is mainly formed in the deoxidation process in the weld pool, for better control of the microstructure of acicular ferrite, it is essential the understanding the nature of the inclusions and the way in which inclusions is formed.

For the nature of the inclusions in the low-alloy steel (Al–Ti–Si–Mn deoxidized steel) weld metal, a lot of research works gave common view. The composition of inclusion could be found on the MnO–Al2O3–SiO2 ternary diagram.2–5) When the concentration (mass%) ratio of aluminum to oxygen is much lower than stoichiometrical value, the central bulk of inclusion show the chemical composition which is on the middle of MnO–Al2O3–SiO2 ternary diagram3,4,8) and it is glassy phase.3,8) On the other hand, when the concentration (wt%) ratio of aluminum to oxygen is approaching stoichiometrical value, the central bulk of inclusion is between gamma-alumina and galaxite in composition.3,8) Furthermore, in both concentration ratio, patch phase of Ti–X and MnS is formed on the surface of the bulk inclusions. There is a possibility that the way in which inclusions is formed is estimated from those nature of the inclusions and the way in which inclusions is formed.

In the present work, toward the understanding of the way in which inclusions may be formed during welding, the inclusion core (primary reaction product in deoxidization process) was directly observed in the weld pool of low-alloy steel, by changing the concentration ratio of aluminum to oxygen from 0.48 to 1.52. Ultra-bright X-ray is used for the direct observation at the synchrotron radiation source. The inclusion formation process is discussed based on those diffraction data and the results of TEM analysis of the inclusion after SAW process.

2. Experimental Procedure

2.1. Weld Metal

The weld metal was prepared by SAW process by changing the combination of wire and flux. The chemical composition of weld metal formed is shown in Table 1. The main difference of chemical composition was the content of aluminum. The concentration (mass%) ratio of aluminum to oxygen for KY1, KY2 and KY4 in Table 1 was changed from 0.48, 0.73 and 1.52, respectively. The microstructure of weld metal is shown in Fig. 1. In the case of KY1 and
KY2, the acicular ferrite was the dominating microstructure. On the other hand, the bainite generating from the grain-boundary was dominating microstructure in the KY4. Those weld metal was used in the Time-Resolved X-Ray Diffraction (TRXRD) experiments for in-situ observation of inclusion core.

2.2. TRXRD Experiments for in-Situ Observation of Inclusion Core

The weld metal prepared includes inclusions which is formed in the deoxidization process of SAW and the inclusion consists of inclusion core and patch phase which is formed at secondary reaction of deoxidization as shown in Fig. 2(a). When re-melting the weld metal prepared by using Gas Tungsten Arc (GTA) plasma, the patch product on the inclusion core disappear due to temperature increase (inclusion core appears) and inclusion is floating and moving in the weld pool as shown in Fig. 2(b). When ultra-bright X-ray irradiates the surface of weld pool in this situation, the moving behavior of inclusion core in the weld pool could be recorded in the reciprocal lattice space. The synchrotron radiation source (SPring-8 in Hyogo, Japan) was used to get the ultra-bright X-ray. The schematic illustration of TRXRD system used is shown in Fig. 3. The weld metal prepared in this research was set on the theta axis of multi-axis goniometer which was at 46XU undulator beam line. The monochromatized X-ray (18 keV) irradiated the fixed point on the weld metal. The incident angle (theta) was 10 degrees. The slit size was 0.1 mm width and 0.5 mm length. The argon arc plasma traverses the fixed point in the constant speeds of 1.0 mm per seconds. The diffracted beam during re-melting the weld metal was recorded on the two-dimensional pixel detector in the time resolution of 0.05 s. The sequence number was assigned to each diffraction patterns corresponding to plasma torch position. The camera dimension was 195 pixels (A pixel has dimensions of 0.172 mm square) in phi direction and 487 pixels in 2-theta direction. The camera was fixed on the 2-theta arm at the 23 degrees. Then the detecting range of 2-theta was from 15.05 and 31.17 degrees. The temperature was roughly estimated by inserting the thermocouple into weld bead and it was related to diffraction patterns by using the sequence number.

2.3. Inclusion Analysis of Submerged Arc Weld Metal

The inclusions in each SA weld metal was sliced by using focused ion beam (FIB). The sliced sample in the FIB device was pasted on copper meshes using a micro sampling device and then thin-foiled. To observe the thin foils, the accelerated voltage of 200 kV was used in TEM. The element analysis of the inclusions was made by EDS.

3. Results and Discussion

Figure 4 shows diffraction patterns recorded on the pixel detector for re-melting KY1 (lower aluminum level). The first image in Fig. 4 is set to 0.0 s for reference (The image sequence number is #872). The horizontal axis correspond to the 2-theta and the vertical axis corresponds to the phi direction shown in Fig. 3. The corresponding temperature was over the measuring range of thermo-couple and it could be said over 2 058 K. There was no diffraction peaks except for the broad and weak halo pattern ranging around 2-theta of 19 degrees. It shows that the number of crystalline oxide is a little. On the other hand, glassy oxide dominates the inclusion populations in much lower aluminum level to oxygen level.

Figure 5 shows diffraction patterns for KY2 (middle aluminum level). The sequence number for the first image in Fig. 5 is as same as that for Fig. 4 for comparison purpose. The corundum alumina was identified and each reflection
index was shown in Fig. 5. At 0.0 s, 116, 214, 300 reflections were identified. However, the reflection index and the position of diffraction pattern along phi direction were changing in time-series as shown in Fig. 5. It corresponded to the moving behavior of inclusion core because the reflection plane which satisfied the Bragg’s law was changing due to motion and rotation of the inclusion core. The results of inclusion analysis after solidification2,5,7) suggests that spinel-type oxide between galaxite and gamma-alumina in composition is dominant phase in inclusion core when aluminun concentration ratio to oxygen is approaching the stoichiometrical value. However, the experiment with KY2 suggested that the corundum alumina and some glassy phase were there in the weld pool.

Figure 6 shows diffraction patterns for KY4 (high aluminum level). The sequence number for the first image in Fig. 6 is as same as that for Figs. 4 and 5 for comparison purpose. The corundum alumina is identified and the diffraction pattern became ring-pattern along phi direction from 0.1 to 0.25 s (116, 024, 113 reflections). It suggests
the number of alumina inclusion is much larger than that for KY2. Then, for concerning the inclusion core in the weld pool, it could be said that the number of alumina increase as increasing the aluminum contents. As shown in Figs. 5 and 6, the corundum alumina was only observed under the arc plasma in GTA process (High shielding-effect). Furthermore, there was no peak corresponding to corundum alumina after arc plasma passed. Those results suggest that the corundum alumina was formed in SAW process for preparation of the samples and it was floated and rotated in re-melting experiment in this time.

Figure 7 shows (a) bright-field image, (b) SAD patterns and (c) EDS mapping for extracted inclusion from SA weld metal of KY1, before re-melting. As corresponding to diffraction patterns in Fig. 4, the glassy phase dominates the inclusion core. The chemical composition of the glassy phase is Mn–Si–Al and it suggests that the phase is on the middle of MnO–Al2O3–SiO2 ternary diagram. Figure 8 is result of inclusion analysis for KY2. In KY2, galaxite is dominant phase in inclusion core and the a little glassy phase was there. It is interesting that in Fig. 5, there is no diffraction pattern corresponding to galaxite. It suggests that the galaxite is formed after solidification. Koseki et al. (18) presented the thermodynamic study of inclusion formation in low alloy steel weld metals. Their calculation results under the thermodynamic equilibrium condition shows mullite (2SiO2·3Al2O3) and Ti3O5 are dominating phase in high temperature and the galaxite is dominating one after solidification in the case of aluminum concentration ratio to oxygen is 0.667. Although the dominating phase in high temperature is different from our experiments, the sequential tendency of inclusion formation is the same. The discrepancy at the high temperature is due to non-equilibrium nature of welding. Figure 9 is results of inclusion analysis: (a) Bright-field image and (b) EDS mapping, for KY4. Al2O3 dominates the inclusion core as expected. It suggests that whole oxygen is killed by aluminum.

The experimental results could be summarized as shown in Fig. 10 along temperature and the concentration ratio of aluminum to oxygen. The image extracted the remaining problem. The possible reaction:
Some glassy phase (in MnO–Al₂O₃–SiO₂ system)

Galaxite ........(1)

should be clear in order to understand the way in which inclusions is formed, in future work.

5. Conclusion

The inclusion core was in-situ observed in the weld pool of low-alloy steel, by changing the concentration ratio of aluminum to oxygen from 0.48 to 1.52. The main conclusions obtained are as follows:

(1) Ultra-bright X-ray and re-melting experiments successfully made inclusion core in the weld pool to be identified. As increasing aluminum contents, corundum alumina was identified as a primary deoxidized product.

(2) In low aluminum contents, glassy phase in Mn–Al–Si–O system dominated the inclusion core as a primary deoxidized product. In middle aluminum contents, corundum alumina plus some glassy phase dominated the inclusion core. The experimental results suggested that the corundum alumina reacted with some glassy phase and the

Al₂O₃+ Some glassy phase
(in MnO–Al₂O₃–SiO₂ system)=Galaxite ........(1)
galaxite was formed. The understanding this reaction is the key for understanding of the way in which inclusions is formed during welding of high strength and low-alloy steel.

(3) For the inclusion formation process, non-thermodynamic equilibrium nature is prominent at the high temperature range of welding. On the other hand, after solidification, the discussion under the equilibrium conditions is effective.

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