Influence of oxide particles on the toughness of modified 9Cr-1Mo steel shielded metal arc weld metals with different Ni+Mn content*

by NISHIKAWA Satoru**, MURAYAMA Shinji*** and NUMAKURA Kotone****

In the modified 9Cr-1Mo steel welds, Ni+Mn content of the filler metal and PWHT temperature range differ between U.S. and Japan. Hence, we investigated the influence of PWHT temperatures on the toughness of modified 9Cr-1Mo steel shielded metal arc weld metals with different Ni+Mn content, it was clarified that the oxide particle density and the hardness of matrix affect the toughness. The weld metal with a high Ni+Mn content of 2.6% partially transformed into austenite during holding on 1073 K which was more than A_{c1} . As a result, the freshly formed martensite of same microstructure as-welded state was formed, the hardness increased and the toughness decreased. As evidence, part of fractured surface after Charpy test is the same as that of as-welded state, spherical oxide particles (almost without any cracks) were observed at the center of the dimple. Therefore, it was suggested that the toughness decreases due to the partial precipitation of freshly formed martensite by carrying out PWHT, and oxide particles affect the decreasing of the toughness.

Key Words: Modified 9Cr-1Mo steel weld metal, Ni+Mn content, Oxide particles, Toughness, post-weld heat treatment, Hardness

1. Introduction

To increase the thermal efficiency and decrease carbon dioxide emissions, it is desirable for coal thermal power plant designers to raise the operating temperature and pressure of boilers. The steam temperature and pressure of ultra-super-critical (USC) thermal power plants are 873 K and 30 MPa, respectively. Such plants require materials with high creep strength. The most prominent example is modified 9Cr-1Mo steel, which is designated as Grade 91 or P/T91. This material, which is developed by modifying the composition of 9Cr-1Mo steel (Grade 9) with minor additions of vanadium, niobium and nitrogen, has much higher creep resistance than 2.25Cr-1Mo (Grade 22) or 9Cr-1Mo (Grade 9) steel. In addition, it has high thermal conductivity and strength, and low thermal expansion. Thus, modified 9Cr-1Mo steel is the most widely used steel in high-temperature parts, such as the super heater and main steam piping¹⁾ of USC thermal power plants.

For Grade 91 or P/T91 base metal, the lower critical temperature (A_{c1}) depends on composition. In general, weld metal may have an A_{c1} temperature ranging 1053 K to 1073 K for Ni+Mn content between 1.0 to 1.5%²⁾, PWHT temperature must be below this temperature. According to ASME B&PV Code Section I 2019, the minimum and maximum holding temperature during PWHT of Grade 91 weld are 978 and 1058 K, respectively. However, the maximum holding temperature can be increased to 1073 K when

Ni+Mn content is less than or equal to 1.0%. In U.S., Grade 91 filler metal which Ni+Mn content is less than 1.0% has been almost used. In Japan, weld metal with Ni+Mn content more than 1.0% is mainly used, according to ministry of economy, trade and industry (METI) standard, the minimum and maximum holding temperature during PWHT are 953 and 1033 K, respectively. Hence, Ni+Mn content of the filler metal and PWHT temperature range differ between U.S. and Japan.

On the other hand, previous reports on the toughness reduction of type 9Cr steel weld metals, include increasing the number of coarse inclusions³⁾, increasing the oxygen^{4), 5)} and carbon content⁶⁾. By contrast, toughness improvement of these weld metals is achieved by the application of high-temperature post-weld heat treatment (PWHT)^{7), 8)}. Also, influence of Ni+Mn content on mechanical properties and microstructure was already investigated⁹⁾. However, the change of microstructure due to PWHT temperature with different Ni+Mn content and how this change affects the toughness of weld metals have not been investigated. In this study, we welded joints using SMAW with different Ni+Mn content, and then subjected these joints to PWHT at different temperatures. We investigated the influence of microstructure on the toughness of the weld metals and determined the dominant factors determining toughness.

2. Experimental procedure

2.1 Materials and welding procedure

The edge shape of the welded joints is shown in Fig. 1. Welded joints were fabricated using SMAW. To suppress the dilution of the chemical composition from the carbon steel plates, the groove

* Received: 2019.11.21, Presented at Visual-JW or WSE 2019

** Member, Department of Physical Science and Materials Engineering, Iwate University

*** Department of Physical Science and Materials Engineering, Iwate University

**** Student member, Graduate School of Arts and Sciences, Iwate University

faces of the welded joints were buttered with three layers of SMAW metal. The welding conditions of each welding processes and chemical compositions of each weld metal are shown in Table 1 and 2. Both weld metals differ to Ni+Mn content (weld metal A of 2.6%, weld metal B of 0.78%) and polarity of welding rod. Subsequently, PWHT was applied to the welded joints for 7.2 ks at 953–1073 K. The heating rate during PWHT was 0.028 K/s and the cooling rate was 0.036 K/s.

Fig. 1 Schematic illustration of the edge shape of the welded joints (unit: mm).

Table 1 Welding condition of SMAW welded joint.

Weld metal	Polarity	Current (A)	Voltage (V)	Welding speed (mm/s)	Number of welding pass	Preheat and interpass temperature (K)
A	AC	150	26	1.73-2.28	12	523-623
B	DCEP					

Table 2 Chemical composition of weld metals. (mass%)

Weld metal	C	Si	Mn	P	S	Ni	Cr	Mo	V	Nb	O(ppm)	N(ppm)
A	0.083	0.41	1.63	0.006	0.0019	0.97	8.68	1.05	0.22	0.028	360	280
B	0.12	0.24	0.68	0.005	0.0034	0.1	8.86	0.97	0.21	0.058	480	350

2.2 Investigation of mechanical properties

We cut 2-mm-deep V-notch Charpy specimens (55 mm×10 mm×10 mm) from the transverse cross-section of the welded joints, with the notch located in the center of the plate thickness. The impact energy and the fracture appearance transition temperature (FATT) were obtained from the Charpy test from 183 to 543 K. At each temperature, the test was repeated on three specimens. In addition, the hardness distribution of the welds shown in Fig. 1 was obtained using the Vickers hardness test at 9.8 N.

2.3 Metallurgical investigation

The specimens for observations were cut from the transverse cross-section of the welded joints. These specimens were given a glossy finish by grinding with metallographic paper and buffing with 1-μm diamond paste, following by etching with 200 ml water and 50 ml HCl and 5 g FeCl₃. Field-emission scanning electron microscopy (FE-SEM, JEOL JSM-7800F) observation of the fracture surfaces after the Charpy test was carried out. In addition,

elemental identification of inclusions was performed using energy dispersive X-ray spectrometry (EDX). The acceleration voltage of the FE-SEM was 15 kV. In the FE-SEM observations, an area of 127.15 μm × 37.54 μm was observed with 10 fields, and the number density of inclusions was measured.

3. Result and discussion

3.1 Toughness of the as-welded weld metals

The impact energy and brittle fracture surface ratio of the as-welded weld metals are shown in Fig. 2 at different temperatures. The fracture appearance transition temperature (FATT) of weld metals A and B were obtained 413 K and 373 K, respectively. Therefore, FATT of the both weld metals are almost same temperature, it showed the same toughness. Fig. 3 shows optical micrographs of as-welded weld metals. Because the martensite lath

Fig. 2 Effect of Ni+Mn content on impact energy and brittle fracture surface ratio of weld metals to the as-welded state.

Fig. 3 Optical micrographs of as-welded weld metals: (a) weld metal A, (b) weld metal B.

of weld metal A is finer than that of weld metal B, the upper shelf energies of weld metal A is higher than that of weld metal B.

The fracture observation results for brittle fracture surface ratios of 0% and 100% are shown in Fig. 4. For a brittle fracture surface ratio of 100%, the fractured surface was flat, with most cracks propagating along the prior austenite grain boundaries. At a brittle fracture surface ratio of 0%, both weld metals exhibited a typical

Fig. 4 SEM micrographs of the fracture surface after the Charpy test of as-welded weld metals.

dimple fracture surface, the dimple size was 3–5 μm . In both weld metals, granular inclusions were observed near the center of many dimples. The sizes of the granular inclusions observed in the both weld metals were similar (0.2–1 μm).

To identify the granular inclusions observed in the both weld metals for a brittle fracture surface ratio of 0%, we carried our EDX analysis (Figs 5, 6). In both weld metals, the granular inclusions (identified by the white arrows) contained a high content of oxygen, manganese and silicon. Hence, we identified these granular inclusions as MnSiO_3 based on a previous study¹⁰⁾. No inclusions other than MnSiO_3 were observed. The oxide particles on the fracture surface were spherical and almost no oxide particles with cracks were observed.

Fig. 5 SEM micrograph fracture surface for as-welded weld metal A. The region at which the spectral pattern was sampled is indicated by the white arrow.

Fig. 6 SEM micrograph fracture surface for as-welded weld metal B. The region at which the spectral pattern was sampled is indicated by the white arrow.

3.2 Toughness of the weld metals subjected to PWHT

The temperature dependencies of the impact energy and brittle fracture surface ratio in the weld metals after PWHT are shown in Figs 7, 8. In both weld metals, the upper shelf energy increases with increasing PWHT temperature. Hence, as reported in previous studies^{7), 8), 10)}, the toughness is improved by increasing the PWHT temperature. Then, as a result of comparing the toughness of weld metals A and B when the PWHT temperature elevated from 1033 K to 1073 K, the toughness of weld metal B was improved, but the contrary result was shown for weld metal A.

Fig. 7 Effect of PWHT temperatures on impact energy by V-notch Charpy test in both weld metals.

Fig. 8 Effect of PWHT temperatures on brittle fracture surface ratio after Charpy test in both weld metals.

The fracture observation results for brittle fracture surface ratios of 0% in the weld metals subjected to PWHT from 953 to 1073 K are shown in Fig. 9. Similar to the as-welded metals in Fig. 4, many dimples were observed on the fracture surface with a brittle fracture surface ratio of 0%. The dimples in the both weld metals fracture surfaces were larger than in the as-welded metals and spherical oxide particles (almost without any cracks) were present in the vicinity of the center of many dimples. However, in weld metal A subjected to PWHT at 1073 K, the dimples of the same small size as the as-welded state were observed on part of the fracture surface (the region surrounded red line). Because the weld metal A with a high Ni+Mn content partially transformed into

Fig. 9 SEM micrographs of the fracture surface for a brittle fracture surface ratio of 0% after the Charpy test of weld metals subjected to PWHT for 7.2 ks at each temperature.

austenite during holding on 1073 K which was more than A_{c1} . As a result, the freshly formed martensite of same microstructure as-welded state was formed, the toughness of weld metal A decreased when the PWHT temperature elevated from 1033 K to 1073 K.

Because the number density of oxide particles decreased as a result of coarsening of the dimples, we measured the oxide particle density on the fracture surface to clarify its contribution to fracture. The effect of PWHT temperatures on the hardness and the oxide particle density observed on the fracture surface with a brittle fracture surface ratio of 0% is shown in Fig. 10 along with the hardness change. When the PWHT temperature was elevated, the oxide particle density on the fracture surface of both weld metals decreased. However, the oxide particle density and the hardness of

Fig. 10 Effect of PWHT temperatures on hardness and oxide particle density of fracture surface for a brittle fracture surface ratio of 0%.

weld metal A was increased by freshly formed martensite when the PWHT temperature elevated from 1033 K to 1073 K, the changes by elevating temperature of the oxide particle density and the hardness showed almost the same tendency. The oxide particles in as-welded state and after PWHT have the same size. Because $MnSiO_3$ is a stable oxide (melting point of 1564 K), we consider that oxide particles do not dissolve in the matrix at 1073 K (i.e., the highest PWHT temperature used in this study). Therefore, the application of higher PWHT temperatures makes it difficult to fracture the oxide particle/matrix interface, thereby reducing the contribution of the oxide particles to fracture. Therefore, because the matrix hardness was reduced by elevating the PWHT temperature, the impact force during the Charpy test on the interface between the oxide particles and the matrix decreased, and the contribution of oxide particles to fracture was reduced. Moreover, the dimples became coarser as the toughness was improved, because the density of oxide particles, which contribute to the formation of dimples, was reduced.

Many carbides were precipitated by PWHT, as shown in Fig. 11; however, almost no carbide was detected on the fracture surface

with a brittle fracture surface ratio of 0%. Hence, we propose that carbides do not contribute to the reduction of toughness. Carbides precipitate from the matrix during PWHT, increasing their coherence with the matrix and thus the interface strength. Here, coherence means that there is a certain orientation between the carbides and the matrix. For this reason, it is considered that the interface between carbides and matrix is difficult to fracture.

Fig. 11 Optical micrographs of the final pass of weld metal A subjected to PWHT at different temperatures.

4. Conclusions

The dominant factors determining the toughness of SMAW metals with different Ni+Mn content are the oxide particle density and the hardness of the matrix. The toughness of weld metal with a Ni+Mn content of 2.6mass% decreased when subjected to the PWHT of 1073 K which was more than A_{c1} . Because it was caused

by the precipitation of freshly formed martensite partially during PWHT.

Reference

- 1) C. R. Brinkman, D. J. Alexander, P. J. Maziasz: Modified 9Cr-1Mo steel for advanced steam generator applications, ASME/IEEE Power Generation Conference, Boston, MA, (1990), 21-25.
- 2) M. L. Santella, R. W. Swindeman, R. W. Reed, J. M. Tanzosh: Martensite formation in 9Cr-1Mo steel weld metal and its effect on creep behavior, Oak Ridge National Laboratory, Babcock & Wilcox Company.
- 3) B. Arivazhagan, S. Sundaresan, M. Kamaraj: Effect of TIG arc surface melting process on weld metal toughness of modified 9Cr-1Mo (P91) steel, Materials Letter, 62 (2008), 2817-2820.
- 4) C. Chovat, E. Galand, B. Leduey: Effect of various factors on toughness in P92 SAW weld metal, Welding in the World, 52, No.7/8 (2008), 18-26.
- 5) B. Arivazhagan, S. Sundaresan, M. Kamaraj: A study on influence of shielding gas composition on toughness of flux-cored arc weld of modified 9Cr-1Mo (P91) steel, Journal of Materials Processing Technology, 209 (2009), 5245-5253.
- 6) B. Arivazhagan, M. Vasudevan: A comparative study on the effect of GTAW processes on the microstructure and mechanical properties of P91 steel weld joints, Journal of Manufacturing Processes, 16 (2014), 305-311.
- 7) R. K. Shiue, K. C. Lan, C. Chen: Toughness and austenite stability of 9Cr-1Mo welds after tempering, Materials Science and Engineering A, 287 (2000), 10-16.
- 8) A. Sharma, D. K. Verma, S. Kumaran: Effect of post weld heat treatment on microstructure and mechanical properties of hot wire GTA welded joints of SA213 T91 steel, Materials Today: Proceeding, 5 (2018), 8049-8056.
- 9) G. Taniguchi, K. Yamashita: Effect of PWHT temperature on mechanical properties of high-Cr ferritic heat-resistant steel weld metals, Kobe steel engineering reports, 63-1 (2013), 84-88.
- 10) S. Nishikawa, S. Okita, Y. Horii: Toughness recovery of modified 9Cr-1Mo steel weld metals after long-term thermal aging and its evaluation by electrochemical measurement, Quarterly Journal of The Japan Welding Society, 26 (2008), 220-226.