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Microstructures and intermediate temperature brittleness of newly developed Ni-Fe based weld metal for ultra-supercritical power plants

Dong Wu, Dianzhong Li, Shanping Lu*

Shenyang National Laboratory for Materials Science, Institute of Metal Research, Chinese Academy of Sciences, Shenyang 110016, China

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ABSTRACT

Temperature dependent tensile behavior of Ni–Fe based weld metal by gas tungsten arc welding (GTAW) was evaluated in the range of 350–820 °C at a strain rate of 0.035/min. Intergranular fracture and intermediate temperature brittleness (ITB) took place at around 750 °C. The microstructure and the fracture surface morphology observation by optical microscopy, scanning and transmission electron microscopy showed that the ITB to a large extent depended on the grain boundary sliding (GBS) and the gamma prime (γ′) precipitation during the elevated temperature tensile deformation. X-ray diffraction (XRD) confirmed that the primary phases formed during the last stage of solidification were mainly TiN nitrides, MC carbides, and Laves phases in the form of Laves/γ eutectics. Quantitative statistics of the Laves/γ eutectics and the MX(MC, TiN) phases were processed. These primary phases had a pinning effect on the migration of grain boundaries and accordingly made the grain boundaries tortuous. The tortuous grain boundaries were expected to inhibit the GBS, relieving ITB. Transmission electron microscopy confirmed that fine Laves particles precipitated along the grain boundary in the weld metal with higher Mo content during the deformation process, and these Laves particles were also supposed to be one factor for inhibiting the GBS.

1. Introduction

Ni-based superalloys, strengthened by solid solution and/or aging, are widely applied in aircraft engine, electric and chemistry industries [1–3]. Ni-based superalloys,e. g. Inconel 617(Ni-Co-Mo based) [4,5] and Inconel 740(Ni-Co based) [6–8] are preferred to be used in the hottest boiler sections, tubes and pipes, of developing 700–760 °C ultra-supercritical (USC) fire power plants due to their better creep strength and oxidation resistance than that of the currently used ferritic and austenitic steels for 600 °C plants [9]. At present, both the superalloys are under development for meeting the demand. In China, GH984 (Ni-Fe based superalloy containing 20% Fe without Co) is considered as a promising candidate alloy for use as boiler material in 700 °C USC power plants due to its lower cost, outstanding workability, evaluate temperature performance and high thermal conductivity [10,11]. For heavy demand for welding application to USC plant boiler tubes and pipes, a kind of Ni-Fe based superalloy welding material is newly developed. The welding material matches with the GH984 alloy in composition and is suitable for welding GH984 alloy. At present, both GH984 alloy and its welding material are under development. However, many Ni based superalloy, including annealed [12–14], cast [15], weld [16,17], directionally solidified [18], single-crystal [19] and even powder metallurgy [20] superalloys, showed low ductility at intermediate temperature. This behavior is known as intermediate temperature brittleness (ITB) and commonly occur between 500 °C and 900 °C [21], which means that remarkable attention to ITB should be paid when it comes to the superalloys serving at around 700 °C in USC plant boilers.

There were many possible explanations for ITB. Roughly speaking, these explanations could be classified into two primary viewpoints. One arose from the change in dislocation movement type in $L1_2\gamma'$ phase in superalloy at intermediate temperature [15,18,19,22] causing an anomalous increase of yield strength and a trough of ductility. The other one was the deterioration of grain boundary strength at intermediate temperature causing grain boundary cracking and a trough of ductility [12,14,16,17,23–28]. Although the studies for the second viewpoint reached a consensus that grain boundaries strength deteriorates at intermediate temperature, the factors causing this deterioration in these studies were quite distinct. High fraction of random grain boundaries [28], grain boundaries with high misorientation angle [25], penetration of gas phases such as oxygen [29] and hydrogen [30], segregation of impurities such as sulfur [27], phosphorus [31] and bismuth [21], and straight grain boundaries [16,17] were all found to deteriorate the grain boundaries strength and led to the intergranular

* Corresponding author.
E-mail address: shplu@imr.ac.cn (S. Lu).

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fracture.

To date, no reports exist on the role of ITB in the new kind of Ni-Fe based welding material, moreover previous studies about the ITB in other superalloys rarely focused on the weld metals, thus, in this work ITB in the new kind of Ni-Fe based weld metal was studied. By means of tensile test between 350 ℃ and 820 ℃ and careful microstructure characterization using scanning electron microscopy (SEM), transmission electron microscope (TEM) and X-ray diffraction (XRD), the mechanism of ITB in the Ni-Fe based weld metal was discussed. In this work, introducing tortuous grain boundaries by adding molybdenum was found to be practicable for improving the ductility at intermediate temperature and inhibiting the ITB of the Ni-Fe based weld metal.

2. Experimental methods

2.1. Material preparation

Two filler metals of Ni-Fe based superalloy, GHHS-1 and GHHS-2, were prepared. The base metal and the backing plate were prepared by GH984 alloy. The chemical composition of the filler metals and the base metal were listed in Table 1. The dimensions of base metal and backing plate were 350 mm×150 mm ×12 mm and 400 mm×20 mm ×12 mm respectively, as shown in Fig. 1.

2.2. Welding process and sample preparation

A orbit welding machine Panasonic TA1600 with a cold wire feeder was employed to prepare the weld metals. Base metal was cleaned with acetone and set a reverse deformation of 15°+15° before clamping. A gas tungsten arc welding (GTAW) was applied in the welding process and the welding current and voltage were controlled in 180 A and 14 V, respectively. The welding speed and wire feeding rate were controlled in an average level of 0.1 m/min and 1 m/min respectively. Argon gas was used as the shielding gas with a flow rate of about 15 L/min. After each pass, oxide scales on the surface of the weld metal were removed with an electric grinder. The interpass temperature was controlled below 100 ℃. The weldment design and tensile test sample dimensions were presented in Fig. 1.

2.3. Tensile test

Tensile tests were conducted in air condition, using a MTS E45. 105 machine with a temperature-controlled furnace at a strain rate of 0.035/min. The samples which had held for 10 min at each test temperature were tested at different temperatures between 350 ℃ and 820 ℃. Elongations of the tensile samples after fracture were measured to evaluate the ductility.

2.4. Microstructural characterization

Specimens for microstructural examination were prepared using standard metallurgical techniques, and finally polished using 2.5 µm diamond pastes. Samples for macrostructure observation of the weldment were etched using a reagent of 10 g copper chloride, 50 ml muriatic acid and 50 ml ethyl alcohol. Samples for dendritic crystal, precipitates and grain boundary observation were electrolytic etched using a reagent of 10 g chromium trioxide and 90 ml water with the voltage of 5 V. Samples for quantitative statistics of the primary phases (MX, Laves phase) were electrolytic etched using a reagent of 10 g sodium hydrate and 90 ml with a current of 0.2 A/cm^2 for 2 s Microstructure and fracture surfaces were examined using scanning electron microscopy (SEM) in a supra 35 microscope equipped with an energy dispersive X-ray spectroscope (EDXS). Area fractions and number densities of the primary phases were statistically analysed by image process software Image J. Considering that the depth of single pass weld was approximately 1.5 mm in our study, a rectangular region of approximately 1.6 mm×75 µm along the depth direction of the weld metal was chosen to represent the entire microstructure of the weld metal. Within this rectangular region at least 14 pieces of SEM images, each of which covered an area of 75 µm×65 µm, were examined to measure the area fractions and the number densities of the primary phases. High magnification observation was carried out using a FEI Tecnai F20 transmission electron microscope (TEM). The TEM foils were prepared by twin-jet thinning in a reagent of 10% perchloric acid and 90% ethyl alcohol at around −25 ℃.

3. Results and discussions

3.1. Microstructure characterization

Fig. 2 illustrated the macrostructure of the GHHS-1 and the GHHS-2 weld metals. The composition differences of the two kind of weld metals didn't cause a significant distinction in the size and growth mode of the grains. Epitaxial growth of the columnar grains was the dominant growth type in the whole weld metal, and the columnar grains grew from the base metal to the top surface of weld metal perpendicular to the welding direction. As the grains growing, the growth direction was parallel to the maximum temperature gradient, meanwhile the grain size became coarser because the orientation preferred grains with [100] orientation grew more rapidly.

Fig. 3(a) and (b) showed that cellular dendrites were the dominant feature within the columnar grains in the GHHS-1 and GHHS-2 weld metals respectively, and the sizes of the cellular dendrites in the two weld metals were nearly the same. Fig. 3(c) and (d) showed the SEM image of the typical dendritic feature of the GHHS-1 and the GHHS-2 weld metals respectively. Primary phases forming in the final stage of solidification process were observed within the interdendritic area, and a larger amount of the primary phases formed in the GHHS-2 weld metal than that in the GHHS-1 weld metal. The X-ray diffraction (XRD) analysis of the precipitates extracted from the GHHS-1 and the GHHS-2 weld metals (Fig. 4) indicated that precipitates in the weld metals contained dominantly (Nb, Ti)C carbides(cubic structure), TiN nitrides (cubic structure) and Laves phases(hexagonal structure) in the form of Laves/γ eutectics. Fig. 5 showed that the blocky or rodlike carbides were almost irregular in shape, and in dimensions of several microns or under 1 µm. TiN nitrides were observed to be nearly blocky and about 1 µm. TiN nitrides always formed during the solidification and MC carbides was also found to start forming in the end of solidification process, and the atomic ratio of Nb and Ti in the MC carbides was variable. As shown in Fig. 5, Laves phases formed in the interdendritic area and was reported to be crystallized with a hexagonal structure and AB_2 stoichiometry [32]. According to the compositional analysis by the EDXS in Table 2, Laves phases observed here can be

Table 1
Chemical composition (wt%) of the filler metals and the base metal (balance Ni).

	Cr	Fe	Mo	Nb	Al	Ti	C	S	P
GHHS-1	21.06	20.6	2.70	3.24	0.43	0.99	0.047	< 0.001	0.004
GHHS-2	20.30	21.0	6.48	2.53	0.35	1.08	0.037	0.0011	0.004
Base metal	20–22	19–21	Sum to 3.1–3.6		Sum to 2.1–2.5		0.03–0.06	≤0.002	0.018–0.022