IOP Conf. Series: Materials Science and Engineering **439** (2018) 052014 doi:10.1088/1757-899X/439/5/052014

Research progress on high temperature materials for 700℃ ultra-supercritical coal-fired units

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Abstract: Nowadays, material research worldwide for 700℃ ultra-supercritical firepower generator units (USC-FPGUs) is focus on the evolution of microstructures and mechanical properties as well as the research progress of high-temperature materials, which are used for fabricating water-cooled wall, superheater/reheater tube, main steam pipe and high-temperature steam header. HCM12 steel and Inconel 617 alloy are suited for high-temperature segment of water-cooled wall. Inconel 740H alloy developed from Inconel 740 alloy after modification is the preferred material for superheater/reheater, main steam pipe and high-temperature steam header due to its excellent persistent creep strength and welding performance. In future research, it is necessary to further verify the high-temperature persistent strength and creep strength of the aforementioned materials through practice. Meanwhile, the optimum hot working process should be exploited to improve the hot-workability and welding performance as well as reduce the cost of materials.

1. Introduction

Since the end of the 20th century, many countries have successively invested a lot of manpower and money to investigate the technology of 700℃ ultra-supercritical coal-fired power generation technology (USC-CFPGT). In Europe, a plan named as ‘The Thermal AD 700 Power Plant’ was launched in January 1998. The target is to develop an advanced ultra-supercritical (A-USC) demonstration power station with the level of 37.5MPa/705℃/700℃, and the thermal efficiency of the units will exceed 50%[1]. Similarly, a plan assigned as ‘760℃ Plan’ was organized and implemented in United States. The objective is to exploit the units with the level of 37.9MPa/732℃/760℃, and the thermal efficiency of the units will also exceed 50%[2]. Moreover, Japanese started ‘New Sunshine’ plan to build A-USC firepower generator units (FPGU). The level and the thermal efficiency of the units are 35 MPa/700℃/720℃ and 48-50%, respectively.

After the localization process of 20 a for China’s FPGU, multiple sets of 600℃ USC generator units have been successfully produced and used in commercial operations. So far, China possesses the largest numbers of USC generator units in the world. It has laid a good foundation for the development of 700℃ USC Coal-fired units. In January 2011, China launched the project of ‘State 700℃ Ultra-Supercritical Coal-Fired Electricity Key Technology and Equipment Research and Development and Application Demonstration’. The goal is to independently complete the overall design for 700℃ USC-CFPG. The level of the units, the capacity and the thermal efficiency would be 35MPa/700℃/720℃, 600 MW and 48-50%, respectively[3]. More importantly, the research and implementation of this key technology will be greatly pushed forward the work of energy conservation and emission reduction in China.

The high-temperature materias used for 700℃ level A-USC CFPUs possesses excellent high-temperature persistent creep performance (the persistent strength ＞ 100MPa at 700℃ and 750℃ for 100000h), long-term stability of microstructures, admirable resistances to steam oxidation inside the pipe and smoke corrosion and fly ash erosion outside the pipeline, easy processing and welding performances as well as lower cost. To provide evidences for the relevant personnel, the authors summarized the research progress of high-temperature materials used for water-cooled wall, superheater/reheater, main steam pipe, outlet header and other components of 700℃ level A-USC units.

2. Water-cooled wall material

When the level of A-USC FPGUs reach 700°C/35MPa, the steam within the water-cooled wall pipe possesses high pressure and temperature, the designed temperature will reach 535-535°C and the highest working temperature will reach 558°C, which requires water-cooled wall material has higher strength and better oxidation resistance. At present, HCM12 steel, Inconel 617 alloy and etc. have been developed for 700°C level water-cooled wall at high-temperature segment. Table 1 shows the chemical composition of HCM12 steel and Inconel 617 alloy.

Table 1. Chemical composition of HCM12 steel and Inconel 617 alloy (mass fraction) %

Grade	C	Cr	Fe	Co	Mo	Mn	Ti	W	Al	Si	P	S	Ni
HCM12	0.10	12.0	Bal.	12.0	1.0	—	—	1.0	—	—	—	—	—
Inconel 617	0.05-0.10	20.0-23.0	≤0.7	10.0-13.0	8.0-10.0	≤0.7	0.2-0.5	—	0.6-1.5	≤0.7	≤0.008	≤0.012	Bal.

HCM12 steel, which is co-developed by Japan's Sumitomo and Mitsubishi Heavy Industry, is 12% Cr (mass fraction, the same below) high alloying elements martensitic steel. HCM12 steel possesses high creep strength, good oxidation and corrosion resistances as well as excellent welding performance. For instance, the water-cooled wall tubes fabricated by this steel does not need heat treatment after welding, but still exhibits excellent service feature. However, δ ferrite with the mass fraction as much as 30% in this steel deteriorates the processing performance. Creep strength will be decimated when the temperature higher than 550°C. Therefore, long term applicability remains to be further studied[4].

Inconel 617 alloy (20% Cr - 50% Ni) is a nickel-based alloy, which is developed in 1970s by original International Nickel Alloy Company (INCO Alloys International) as a high-temperature alloy strengthened by Cr-Mo-Co solid solution and hardened by carbide[5]. The addition of Al and Ti promotes the formation of a small amount of γ'-Ni3 (A l/Ti) phase with the amount of 4-5%, which can cause precipitation strengthening. Also, a small amount of M6C and Ni2(Cr, Mo) phases are formed in the aging treatments. No σ (affecting the high temperature strength and corrosion resistance, thus affecting the service life) and μ phases are detected in the aging treatments. Therefore, Inconel 617 alloy has been incorporated into the ASME Code Case 1956 standard case, which can be acted as an alternative material for HCM12 steel. More importantly, Inconel 617 alloy also has been selected as the first boilers material for Europe A-USC plan. Compared to HCM12 steel, Inconel 617 alloy has higher strength and better ability to resist oxidation corrosion, but the price is nearly 10 times than HCM12 steel, and manufacturing cost is also high. It is noted that T23 steel, T24 steel and T92 steel are as low temperature segment alternative materials in water-cooled wall material selection schemes of 700°C level A-USC units in United States and Europe due to their high-temperature strength and oxidation corrosion resistance are lower than those of HCM12 steel and Inconel 617 alloy.

3. Superheater material

The superheater/reheater is arranged in the smoke area of boiler with the highest temperature. The temperature of the tube wall is higher than that in the medium within the tube by 20-90% when in the working conditions. Due to the long-term effect of high-temperature stress, the material of superheater/reheater not only bears larger creep, but also bears the corrosion and abrasion of high-temperature flue smoke. 700℃ level A-USC units not only require enough high creep strength, persistent strength and plasticity, but also demand superior stability of microstructure in the long time working at high temperature, good welding and processing performances and high steam oxidation resistance (oxidation rate < 0.1 mm a-1). The extensively used 9-12% Cr martensite heat-resisting steel and traditional austenitic stainless steel for 600℃ USC units cannot meet the current requirements. Therefore, nickel-based or iron-nickel-based high-temperature alloys with better high-temperature performance become the preferred materials[6]-[7][8].

American Electric Power Research Institute (EPRI) has carried out a detailed study on the persistent strength of the current used USC generator units. The corresponding results are summarized and exhibited in Figure.1. The chemical compositions of different materials are listed in Table 2. Std. 617 in Fig. 1 is the Inconel 617 alloy, which will not be repeated in this paper. From Fig. 1, it can be seen that CCA617, Nimonic 263 and Inconel 740 nickel-based high-temperature alloys can satisfy the persistent strength requirement for the superheater/reheater tubes of 700℃ level A-USC units, in which Inconel 740 alloy performs the best.

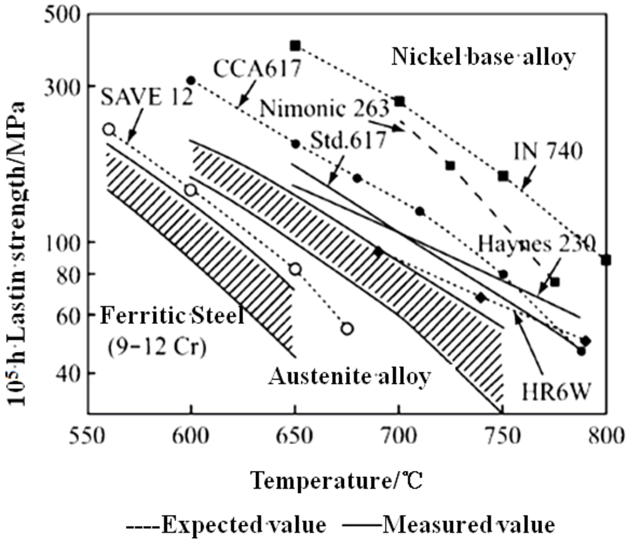

Figure 1. Comparison of creep rupture strength among candidate materials for USC units superheater

Table 2. Chemical composition of materials for superheater/reheater tube operating at 700℃ (mass fraction) %

Grade	C	Si	Mn	Co	Cr	Fe	Mo	Nb	Al	Ti	S	P	Cu	Ni
Inconel 617B	0.05-0.08	≤0.3	≤0.3	11.0-13.0	21.0-23.0	≤1.5	8.0-10.0	≤0.08	0.8-1.3	0.25-0.5	≤0.008	≤0.012	≤0.5	Bal.
Nimonic 263	0.04-0.08	≤0.4	≤0.4	19.0-21.0	19.0-21.0	≤0.7	5.6-6.1	—	≤0.6	1.9-2.4	—	—	≤0.2	Bal.
Inconel 740	0.03	0.05	0.3	20.0	25.0	0.7	0.5	2.0	0.9	1.8	—	—	—	Bal.
Inconel 740H	0.03	0.15	0.3	20.0	25.0	0.7	0.5	1.5	1.35	1.35	—	—	—	Bal.
GH4700	0.033	—	—	20.0	25.0	—	—	—	1.35	1.61	0.0006	—	—	Bal.
AFA	0.07	0.15	2.0	—	14.0	Bal.	2.46	0.85	2.25	—	—	—	—	20.0

CCA617 alloy, which is known as Inconel 617B alloy, is developed by Germany company VDM on the basis of composition optimization of Inconel 617 alloy (lower the upper limit of C, Cr, Fe, Mn and Al, narrow the scope of components). It is a new nickel-based alloy strengthened by solid solution and a small amount of γ' phase. The creep strength at 700℃ for 100000h of this new alloy is higher than that of Inconel 617 alloy by about 25%[9].

In Inconel 617B alloy, the higher contents of alloying elements of Ni and Cr lead to strong corrosion resistance, and the co-effects of Al and Cr result in good high temperature oxidation resistance, Co and Mo cause the effect of solid solution strengthening and make it easy for forming and welding. The persistent strengths of Inconel 617B alloy at 700℃ and 750℃for 100000h are predicted to be 120MPa and 90-100MPa, respectively. It is noted that the experimental data of persistent strength under 30000h have been obtained in Europe. Due to the high content of Mo, the corrosion resistance of flue gas and sedimentary sulfate corrosion resistant ability of Inconel 617B alloy are relatively weak[10]. Therefore, this alloy should be solution treated before used, in which coarse grain structures would cause high creep rupture strength. The superheater tube made of Alloy617B is shown in Figure.2.

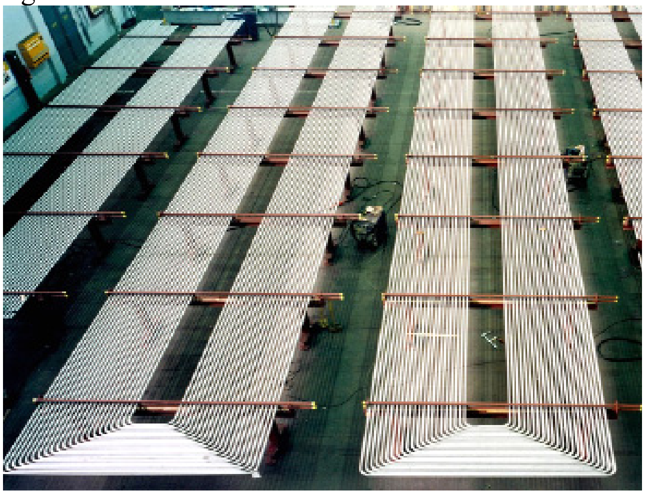

Figure 2. Superheater made by Alloy 617B

Nimonic 263 alloy, which is developed by British Rolls-Royce company in 1971, is a kind of nickel-based alloy relies on γ' phase hardening and solid solution strengthening[11]. The main effect of Mo in the chemical composition is solid solution strengthening. About 6% of γ' phase formed by the precipitation of Ti and Al leads to precipitation strengthening. A small amount of MC and M23C6 carbides and η phase can be detected at high-temperature ageing treatments. The predicted persistent strengths at 700℃ and 750℃ for 100000h are 150MPa and 115MPa, respectively. However, the corrosion resistance is poorer and preparation is difficult for this alloy[12].

Inconel 740 alloy, which is developed by American Special Metals Corporation, is the aging precipitation strengthening type nickel-based alloy. It is one of the candidate materials for 700℃ level FPGU since its high potential allowable stress[13]. Inconel 740 alloy is a new nickel-based alloy on the basis of Nimonic 263 alloy by reducing the content of C and Mo to about 0.04% and 0.5%, respectively, increasing the content of Cr to 24%, and adding 2% Nb. This alloy, which is mainly rely on γ' phase strengthening, is the new type high temperature resistant material used for superheater/reheater in Europe's 'Thermal AD 700 Power Plant' and Germany's 'MACRO DE2' programs. The predicted persistent strength at 700℃ and 750℃ for 100000h are 210MPa and 111MPa, respectively. The oxidation corrosion depth ≤ 2 mm after 200000h. Both of strength and corrosion resistance of this alloy are better than those of Nimonic 263 alloy. As-supplied state of Inconel 740 alloy is solid solution treatment and the solution treatment process is conducted at 1150℃ for 30 min and followed by water quenching and then aging treated at 800℃ for 16h. The microstructure is single-phase austenite accompanied by a large number of twins. The grain size is not uniform (10-120μm) and the average grain size is about 50μm.

Xie et al[14]. demonstrated that Inconel 740 alloy would show the microstructure instability after long-term aging at high temperature. This is because that γ' strengthening phase grows up, η phase precipitates in intragranular and intergranular, the formation of G phase at grain boundary makes it appear instability. Below 725℃, the growth rate of γ' phase in the alloy is slow, the formation of η and G phases is also less and the microstructure stability is good. Above 725℃, acicular η phases nucleate at the grain boundary and grow into the intracrystalline and form γ' phases and then grow up rapidly. Meanwhile, Si provides a stable formation of G phases at the same time. Therefore, stability of microstructures at high temperature is poorer. By improving Al content, decreasing Ti content and strictly controlling the Si content of the composition Inconel 740 alloy, Inconel 740H alloy is obtained and its nominal chemical composition is shown in Table 2. Stabilities of microstructure and of γ' phases in this alloy are greatly improved. More importantly, η and G phases would not appear after aging at 750℃ and 800 ℃ for 5000h. The SEM microstructures of the Inconel 740 alloy heated at 750℃ for 4000h and Inconel740H alloy heated at 750℃ for 5000h is shown in Figure.3

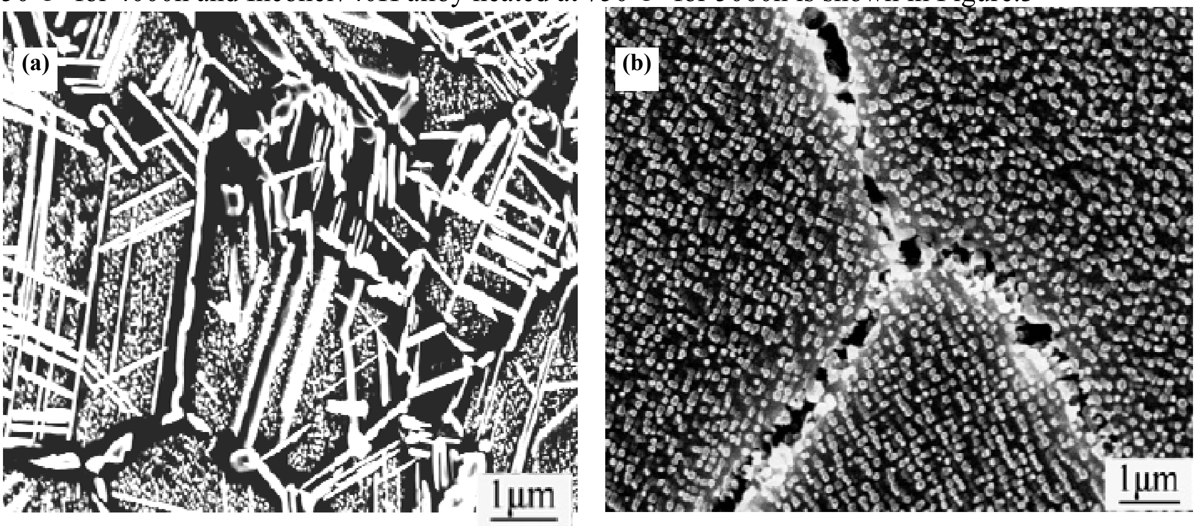

Figure 3. SEM micrography of Inconel 740 and Inconel 740H (a) Inconel 740, (b) Inconel 740H

Zeng et al. design GH4700 nickel-based alloy on the basis of Inconel 740 alloy. The nominal composition is 55Ni-25Cr-20Co, which is listed in Table 2. Hot workability of GH4700 alloy is poor due to the existences of harmful impurity elements segregation and low melting eutectics. This alloy is extremely sensitive to the deformation temperature and strain rate. With the decrease of deformation temperature and the increase of strain rate, the flow stresses rapidly increase. Above research is of theoretical significance for further studying the microstructure evolution, understanding the thermoplastic characteristics and determining the process parameters of in the hot deformation.

Yamamoto et al. developed a new type of austenitic heat resistant steel containing Al on the basis of austenitic heat resistant steel by regulating the element contents, such as Ni, Al, Nb and C[15]-[17]. This steel is strengthened by nanoscale NbC precipitate and has the ability to form Al protective film by itself (Al2O3-Forming Austenitic Stainless Steel, AFA). AFA steel has the same high temperature persistent strength as nickel-based high-temperature alloy. Meanwhile, it has excellent steam corrosion resistance and processing performance. Its typical chemical composition is shown in Table 2. Compared with other new austenitic heat resistant steel (such as NF709 steel) and Inconel 617 nickel-based alloy, AFA steel contains a relatively small number of precious metal elements, such as Cr, Ni, Co and Mo. Therefore, the price is relatively cheap. This kind of new type aluminized austenitic heat resistant steel is expected to be as heat-resisting material for high-temperature components of 700℃ level A-USC units. Currently, researches on such materials are being carried out by Oak Ridge National Laboratory of American, Alstom of France, Metal Institute of Chinese Academy of Sciences and Shanghai Jiaotong University of China.

4. Main steam tube and outlet header materials

Main steam tube and outlet header thick wall tube are mainly used to convey high temperature and high pressure steam, and their structures are relatively complex. Since there are lots of intubation in the walls, enough high creep strength, persistent strength, persistent plastic and thermal fatigue resistance are required. Meanwhile, good fracture toughness, welding and machining performances are necessary. The contacts of the steam tube surfaces need to have good steam oxidation resistance. When the steam temperature up to 700℃, Nimonic 263, Incone l617B and Inconel 740 alloys should be adopted[13][18]-[19].

A major challenge for successful application of A-USC technology is to achieve commercial scale production of the major components of units, relating to smelting, prevention and control of segregation in large castings, making suitable hot working process to avoid the material fracture in the forging and rolling processes, etc. At present, Wellman Gordon Incorporation has successfully developed header pipe used Nimonic 263 alloy with the diameter of 378mm, thickness of 88mm, length 8.9m for Europe A-USC project[13]. The persistent strength of this alloy is high and the performance is close to that of Inconel 740 alloy. The wall thickness is reduced when the alloy used as large diameter pipe at 700℃, but the processing production of the large diameter pipe is difficult.

In addition, V&M Company of France has successfully fabricated large diameter thick wall pipes used for high-temperature pipelines and header by exploiting Inconel 617B alloy. The production processes are as follows: vacuum induction melting - electroslag remelting - forging and preheating - hot forming – rushing and pulling - final molding on the pilger roll mills - solid solution annealing treatment. Through the above process, thick wall tube of Inconel 617B alloy with the diameter of 330-420mm and wall thickness of 25-80mm can be processed. The creep strengths of above pipes at 700℃ and 725℃ for 100000h are 140MPa and 110MPa, respectively. In addition, experiments similar to prevent weld stress relaxation in the fracture of the annealing treatment after welding of Inconel 617B alloy thick wall pipe are carried out[19]. The results show that the creep resistance not significantly reduced, indicating that this alloy has good welding performance in some extent. Low strain rate tests show that the welding residual stress leads to fracture problems of Inconel 617B alloy can be prevented through heat treatment at 700℃ for 3h after welding and cold deformation. Also, slower cooling to room temperature after welding can effectively improve the toughness of the welding line[9]. The superheater header made by Alloy 617B is shown in Figure.4.

Figur 4. Superheater header made by Alloy 617B

Young et al. studied the welding of Inconel 617B alloy under various atmosphere and parameters[20]. The results suggest that argon tungsten-arc welding (TIG) using Tissen 617 wire under the protection of mixed gas Ar+2.5%H2 (volume fraction) can ensure welding joints obtain the optimal microstructures and performances. Moreover, there is no obvious difference in microstructure and composition between matrix and heat affected zone (HAZ). However, the microstructure in the welding line is relatively coarse. The tensile strength (TS) ratio between welding line and matrix is 0.89 and the yield strength (YS) ratio is 0.96 at room temperature, while the TS ratio is 0.86 and the YS ratio is 0.71 at high temperature of 700℃. This result indicates that Inconel 617B alloy has good welding performance and the loss of strength of the welding line is less in the welding process.

In 2011, United States Special Metal Company produced the largest Inconel 740H alloy ingot with the diameter of 750 mm by vacuum induction smelting (VIM) and vacuum arc re-melting (VAR)[21]-[22]. No macro segregation is detected in cross section structure. Carbide and bulky γ' phases are observed while no η phase is detected in microstructure (Figure.5a). After upset forging, punching and extrusion, the ingot can be made into large diameter pipes with the diameter of 380mm, the wall thickness of 89mm and the length of 10m, which can be used to fabricate header and the main steam pipe and other large diameter thick wall pipes (Figure.5b). More than 30000h creep rupture test data indicate that Inconel 740H alloy pipes possess excellent tensile and yield strength in the operating conditions within 760℃.

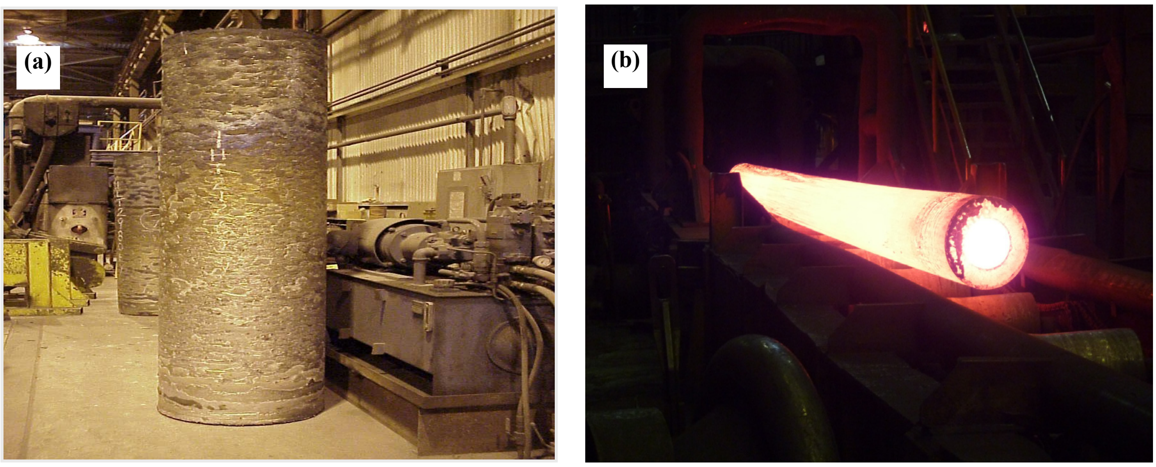

Figure 5. Big cast ingot and thick wall pipe of Inconel740H (a) *Φ* 750mm (b) OD380×89mm×10000mm

Due to the high content of alloying elements of Inconel 740 alloy, liquefied crack is easily formed at the grain boundary in the HAZ of the thick wall tube when welding (Figure.6a). However, the welding performance is greatly improved in the thick wall tube fabricated by the modified Inconel 740H alloy, in which liquefied crack and other defects are not easy to be formed in the welding line and in the HAZ (Figure.6b).

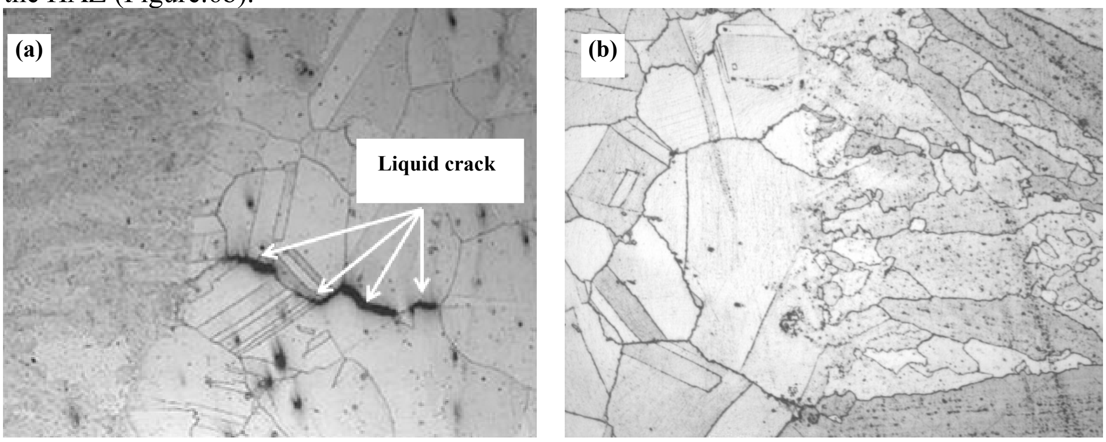

Figure 6. Comparison of weld microstructure of Inconel740 (a)and Inconel740H (b)

As shown in Figure.7, the Inconel 740H alloy header tube butt welded joint with specification of Φ380mm×72mm has been produced by using the hot wire gas tungsten arc welding (GTAW). Before welding, solid solution treatment at 1120℃ and then ageing treatment at 800℃ for 4h should be firstly done. Groove with 5° and the filler wire with Φ0.9mm are needed. After welding, ageing treatment at 800℃ for 4h is completed in the welding line. Test results show that there are no cracks and other discontinuity defects in welding lines, indicating that thick wall pipe of Inconel 740H alloy has good welding performance.

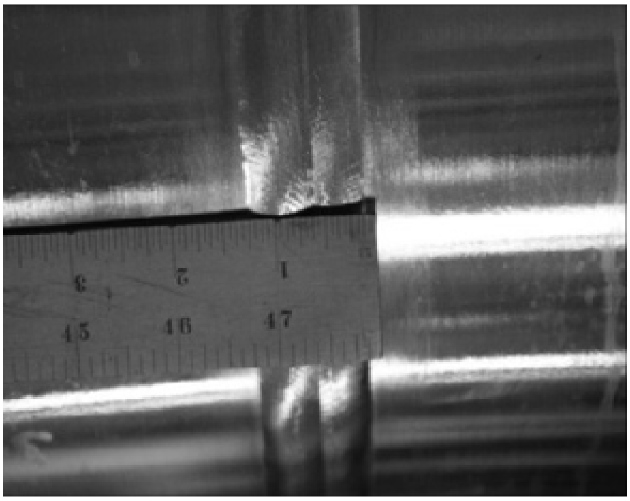

Figure 7. Butt weld of Inconel 740H header

5. Conclusions

Due to the water-cooled wall of 700℃ level A-USC units is served at high pressure and temperature, HCM12 steel and Inconel 617 alloy are selected as the high-temperature segment materials since both of them have excellent strength, admirable oxidation resistance and corrosion resistance at high temperatures. Inconel 617B alloy is developed from Inconel 617 alloy by strictly controlling the content of alloying elements. The high-temperature creep strength of Inconel 617B alloy was higher than that of Inconel 617 alloy by 25%. Therefore, Inconel 617B alloy can be acted as a candidate material for medium and low temperature superheater/reheater tubes. Inconel 740H alloy, based on the Inconel 740 alloy by tightly controlling the content of alloying elements, have better high-temperature stability of microstructures and better long time durability at high temperatures, which can be acted as the best candidate material for the superheater/reheater tubes when the temperature of water-cooled wall is about 750℃. For the materials used for main steam pipe and high-temperature outlet header, Europe and the United States have fabricated thick wall pipes and high-temperature headers by using Inconel 617B alloy and Inconel 740H alloy, respectively. Both of them have good welding performances. However, based on the existing data, predicted persistent strength of Inconel 740H alloy at 750℃ for 100000h is higher than that of Inconel 617B alloy by 10%. Therefore, Inconel 740H alloy should be the preferred material for the main steam pipe and high-temperature outlet header.

Lots of work has been carried out on the high-temperature materials used for 700℃ USC FPGU and much high quality progress has been made. However, the hot working and welding performances of the above mentioned high-temperature materials should be further researched and improved if they are prepared for production practice on a large scale. Also, high-temperature persistent strength and high-temperature creep strength and other performances also need to be further verified by practice. All of these aspects are also the focus and the difficulty in future research.

Acknowledgements

This research was financially supported by Science and technology project of Inner Mongolia Electric Power Research Institute (Grant No. 2017-10).

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