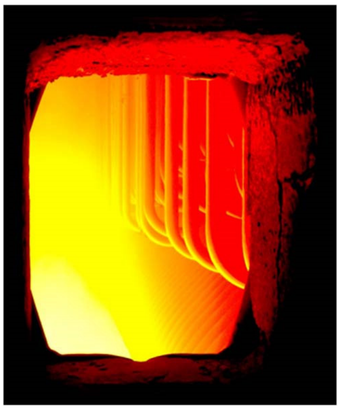
Development of Materials for Use in A-USC Boilers



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The 700°C-class advanced ultra-supercritical (A-USC) boiler is a promising technology for high-efficiency coal-fired thermal power generation. To realize such boilers, it is critical to develop and validate the creep enhanced Ni-based alloys and advanced ferritic steels, both of which can withstand temperatures substantially higher than conventional boilers. In addition to the long-term creep rupture strength and manufacturing techniques, the steam oxidation properties and the coal ash-induced high-temperature corrosion properties were evaluated. The results indicate that the test materials have sufficient strength and corrosion resistance and the pressure parts can be manufactured without problems. The heat transfer tube panels and pipes were additionally installed on existing boilers in May 2015. Since then, the demonstration operation has been continued in a simulated A-USC boiler environment. We will also develop technologies for creep damage assessment and non-destructive inspection for maintenance purposes, aiming for the early commercialization of A-USC boilers.

1. Introduction

From the perspective of reducing the CO_2 emissions and fuel cost of power generating facilities, the "Technological Road Map for Next-generation Thermal Power Generation"[1] was compiled at the consultative meeting held by the Ministry of Economy, Trade and Industry (METI). It provides the guidelines for the lower environmental load and the increased efficiency of power generating facilities. In coal-firing power generation, A-USC boilers with an increased steam temperature of 700°C are considered to be the key technology for the realization of better efficiency. Under such circumstances, a project for the development of 700°C-class A-USC boilers, which is subsidized by METI, has been progressing in Japan since fiscal 2008 (hereafter referred to as the national project). The participants in this project are the major manufacturers and research institutions in Japan (IHI, Toshiba, Fuji Electric, Nippon Steel & Sumitomo Metal, ABB Bailey Japan, Okano Valve Mfg., Toa Valve Engineering, National Institute for Materials Science, Mitsubishi Hitachi Power Systems, and Mitsubishi Heavy Industries) and the cooperation among these entities propels the development. We have been actively involved in the project and are responsible for the evaluation of A-USC boiler material properties and verification of manufacturing. The heat transfer panels and pipes were jointly installed on existing boilers, for demonstration in a simulated A-USC boiler environment. This report introduces our development and the evaluation results of A-USC boiler materials.

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2. Outline of A-USC boilers

Coal is an energy source with abundant reserves. The A-USC boiler is a steam power generation technology using it as a fuel. The final steam temperature of A-USC boilers is 100°C higher than that of conventional coal-fired boilers. Their basic configurations remain unchanged. As there are also no drastic changes required in the peripheral equipment of a power station, an A-USC boiler can be retrofitted to an existing power generating facility as-is in most cases. Therefore, it is expected to be useful as a replacement for aging coal-fired boilers, which are rapidly increasing in number. Because A-USC boilers can also be easily applied to overseas coal-fired thermal power plants, this is one of the technologies that realize the effective use of coal and enable us to contribute to CO_2 reduction worldwide.

Figure 1 compares CO_2 emissions from 1,000MW coal-fired power plants.[1] In A-USC power generation, CO_2 emissions can be reduced by approximately 12% from existing ultra-supercritical (USC) power generation. In A-USC, the efficiency is also expected to be improved by nearly 10% (as a relative value) from USC, which indicates the possibility of maintaining high efficiency when combined with carbon dioxide capture and storage (CCS) systems in the future. The ultra-high temperature materials that are developed can be applied to not only coal-fired power generation, but also the bottoming cycles (steam turbines and waste heat recovery boilers) of gas turbine combined cycle (GTCC) plants with ever increasing temperatures. The extent of their applicability is also the subject of attention.

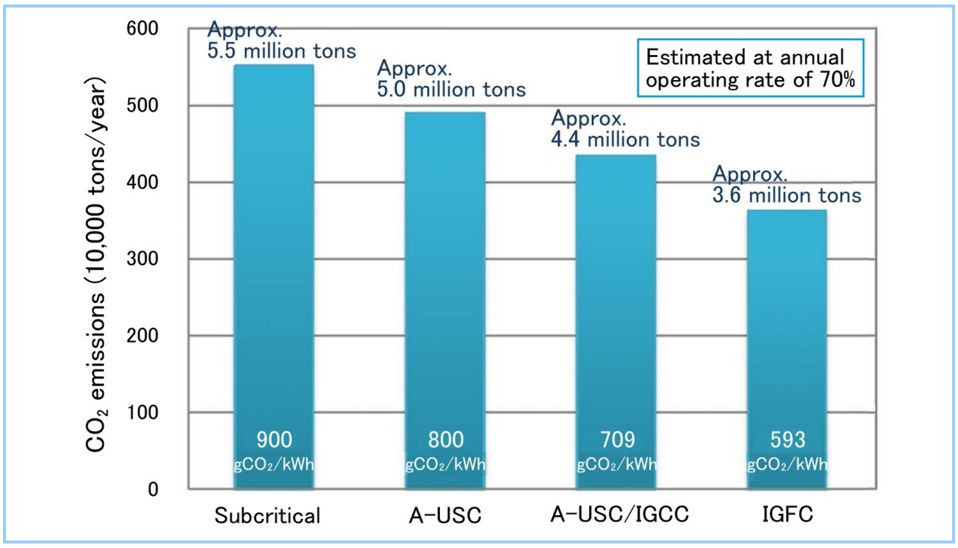

Figure 1 Comparison of CO_2 emissions from 1,000MW coal-fired power plants[1]

Figure 2 is a diagram of a basic A-USC plant configuration.[2] **Figure 3** shows the materials constituting the boiler.[3] Because of their high-temperature strength properties and corrosion resistance, Ni-based alloys with superior heat resistance are intended to be used in heat transfer tubes, main steam pipes and reheat steam pipes, all of which are subject to steam temperatures of 700°C or higher. To improve the economy by decreasing the amount of expensive Ni-based alloys used, advanced ferritic steels are to be used in the major piping with steam temperatures of 650°C or below, whenever possible.

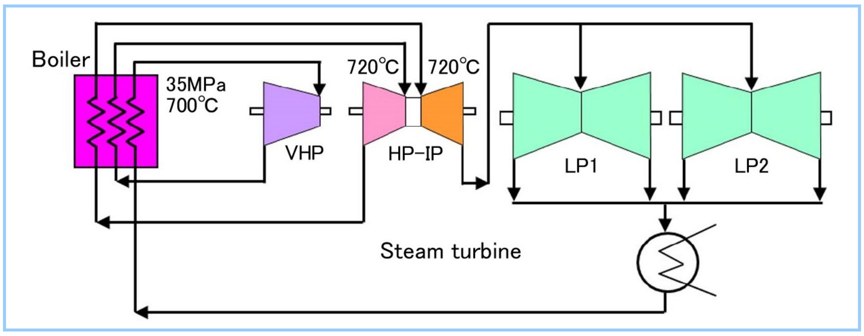

Figure 2 Diagram of the basic A-USC plant configuration[2]

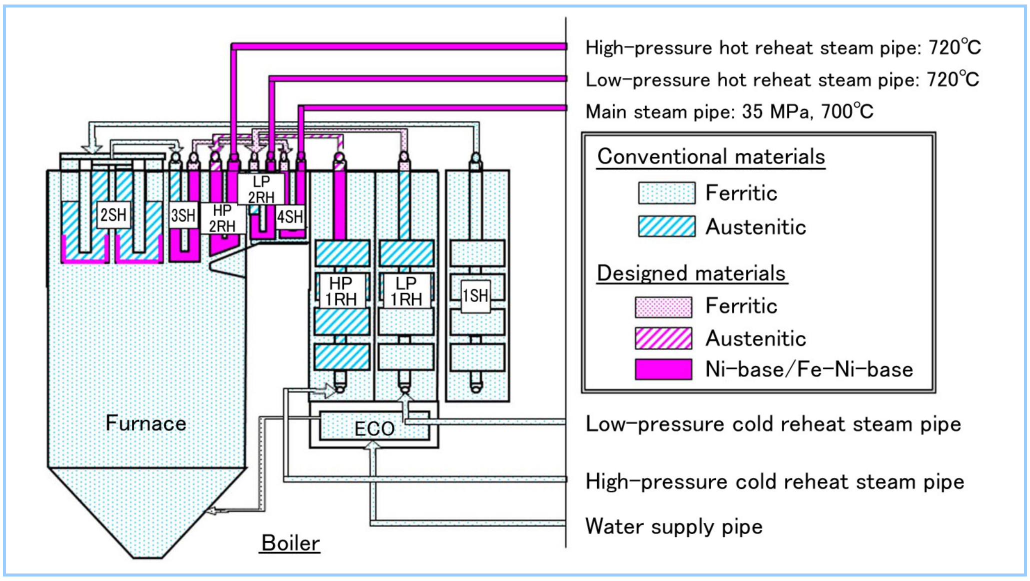

Figure 3 Materials that constitute the boiler[3]

To realize A-USC boilers, therefore, it is important to design/examine Ni-based alloys and advanced ferritic steels suitable for use at temperatures above 600°C and establish manufacturing techniques.

3. Development of boiler component technologies

To select the materials that can be used in A-USC boilers ("candidate" materials), we developed techniques for the manufacturing of pressure parts as boiler component technologies and evaluated the creep strength and corrosion resistance.

3.1 Candidate materials

Table 1 lists the candidate materials for use in A-USC boilers. Six types of Ni-based alloys were included in total: domestically-developed HR6W,[3] HR35[4] and Alloy 141[5] as well as Alloy 617, Alloy 263 and Alloy 740, the properties of which were well assessed in the U.S. and Europe as A-USC candidate materials. Of these, HR6W, HR35 and Alloy 617 have superior workability and high-temperature strength, and are considered especially suitable for use in thick large-diameter pipes. For heat transfer tubes, in addition to these three alloys, Alloy 263, Alloy 740 and Alloy 141 are also included as candidate materials. Their high strength is enhanced by the increased precipitation of γ' phase (Ni_3(Al, Ti) intermetallic). For advanced ferritic steels, the following three types are included: high B-9Cr steel, low C-9Cr steel and SAVE12AD. All of them were developed domestically and were designed by adding Co and/or B to the conventional material (modified 9Cr-1Mo steel) in order to improve the high-temperature strength and prevent deterioration of the creep strength at the heat affected zone (HAZ). **Figure 4** shows the appearances of the test materials used in the various tests. Most of the plates, small-diameter tubes and large-diameter pipes were prepared by material manufacturers in Japan, while the small-diameter tubes of Alloy 617, Alloy 141, Alloy 263 and Alloy 740 were procured from overseas material manufacturers.

Table 1 List of A-USC boiler candidate materials

Material name		Composition	Thick large-diameter pipe	Small-diameter tube
HR6W	Ni-based	45Ni-23Cr-7W	- High-temperature header and connecting pipe - Main steam pipe - Hot reheat steam pipe	- Hot heat transfer tube
HR35	Ni-based	50Ni-30Cr-4W-Ti		
Alloy 617	Ni-based	Ni-22Cr-12Co-9Mo-Ti-Al		
Alloy 263	Ni-based	Ni-20Cr-20Co-6Mo-2Ti-Al	-	
Alloy 740	Ni-based	Ni-25Cr-20Co-2Nb-2Ti-Al	-	
Alloy 141	Ni-based	Ni-20Cr-10Mo-2Ti-Al	-	
High B-9Cr steel	Advanced Ferritic steel	9Cr-3W-3Co-Nb-V-B	- Header - Connecting pipe (up to ≈650°C)	- Heat transfer tube (Temperature range similar to conventional high Cr steel)
Low C-9Cr steel	Advanced Ferritic steel	0.035C-9Cr-2.4W-1.8Co-Nb-V		
SAVE12AD	Advanced Ferritic steel	9Cr-3W-2.6Co-Nb-V-B		

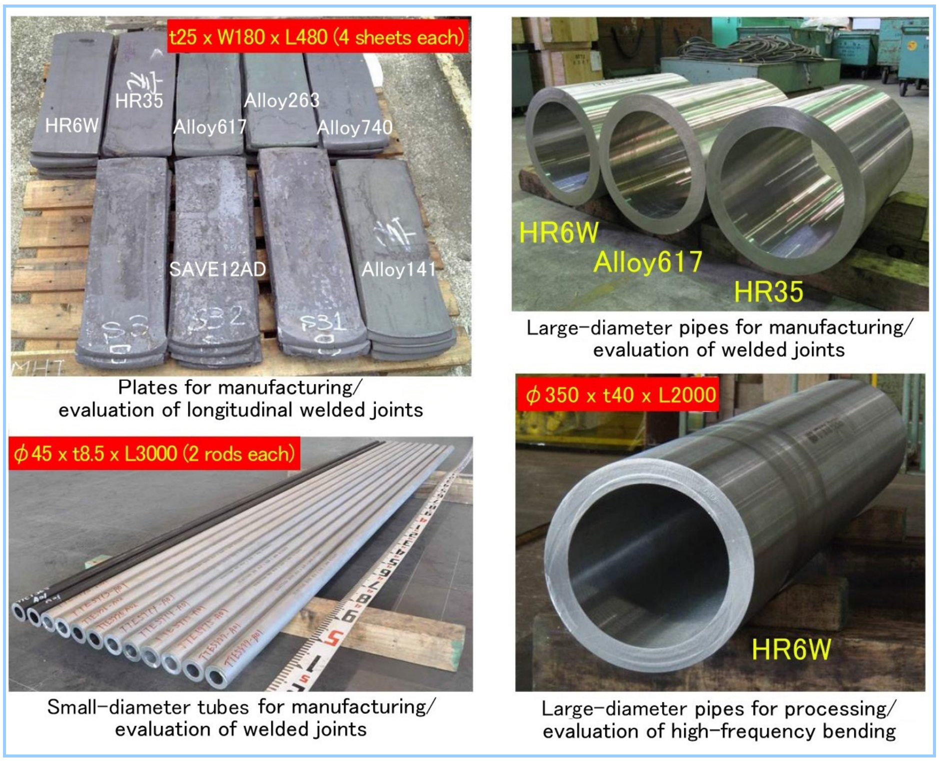

Figure 4 Appearances of the test materials

3.2 Establishment of manufacturing techniques

(1) Welding technique

The automated narrow-groove gas tungsten arc (GTA) process was used to weld small-diameter pipes. The composition of the weld material was similar to the base metal. For the examination of material properties, the Ni-based alloys were used as welded, whereas advanced ferritic steels were thermally treated after welding. The structures of the welded joints were metallographically normal, and no abnormalities were seen in the hardness distribution. The mechanical properties were generally good and were comparable with the base metal.

For the large-diameter pipes of the three alloys, the automated narrow-groove GTA process was applied for welding. However, considering the possibility of on-site welding, HR6W was welded using the shielded metal arc process. The dimensions of the large-diameter pipes used in testing are: outside diameter (φ) 350 mm x wall thickness (t) 40 mm. The photographs in **Figure 5** are HR6W large-diameter pipes in all-position automated GTA[6] welding or shielded metal arc welding. The cross-sectional examination of post-welding structures indicates the presence of micro cracks at HAZ of the Alloy 617 welded joint. These cracks are considered to be hot cracking because of their forms and time of occurrence. With help from material manufacturers, we lowered the content of the trace constituents in the alloy such as boron. The welding conditions were also optimized, considering the welding heat input. As a result, we succeeded in preventing hot cracking of the Alloy 617 welded joints at the next trial.[7] For each test material, the properties of the welded joints were good, indicating the practical applicability to the actual units.

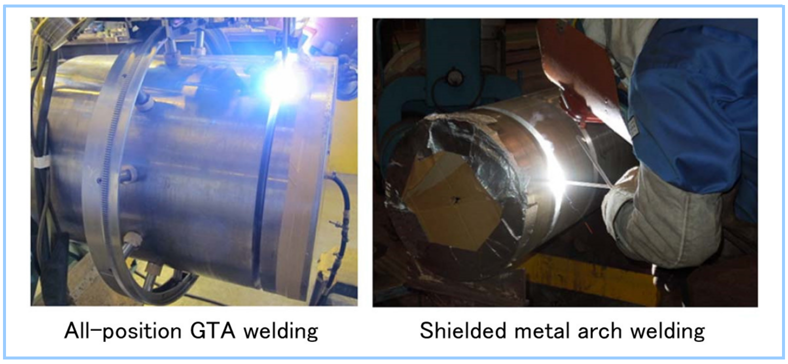

Figure 5 Welding of the HR6W large-diameter pipes

(2) Bending technique

Because the manufacturing of boiler superheaters and reheaters panel involves various types of bend processing, the test materials were also bent according to the existing standard procedure. The bend processing referred to here is of two types: cold bending and hot bending. The bend radius of these bending types is 2.5D and 1.5D, respectively (where D is an outside diameter of the tube). After bending, re-solution heat treatment was conducted on the Ni-based alloys under the same conditions as the original tube, while the advanced ferritic steel (SAVE12AD only) was thermally treated again by normalizing and tempering.

No post-bending abnormalities were found in the outer surface and the workability was good. The flattening of the cross section of the tube at the bend apex and wall thickness decrease rate at the extrados were within the range that is generally expected by the bending of boiler steel tubes, thus causing no problems. The microstructure and hardness at the extrados, intrados and neutral axis were almost the same as the original tube, and the obtained results were considered satisfactory.

In the bend processing test of large-diameter pipes (φ 350 mm x t 40 mm), thick wall steel pipes of Ni-based alloys (HR6W and Alloy 617) and advanced ferritic steel (SAVE12AD only) were used. The bending test was conducted using the same method as conventional steel. Specifically, the pipe was heated locally in a circle with a high-frequency induction heater, while it was pressed/bent by the roller. **Figure 6** is the photograph of bend processing of HR6W large-diameter pipes (bend radius: 4D). After bending, re-solution heat treatment was conducted on the Ni-based alloys (HR6W and Alloy 617) under the same conditions as the original pipe, while the advanced ferritic steel (SAVE12AD) was thermally treated again by normalizing and tempering. The material properties were then examined. For each test material, the properties were almost as good as those of the original pipe, thus demonstrating that appropriate post-bending heat treatments can realize properties equivalent to the original pipe.

(3) Mock-up fabrication

To verify the developed manufacturing techniques, we used HR6W to fabricate mock-ups of the superheater outlet header, the reheater outlet header, and the superheater heat transfer tube panel. As components of a 700°C-class A-USC boiler, the outlet header dimensions were determined to be φ 558.8 mm x t 138 mm for the superheater and φ 635 x t 72 mm for the reheater, respectively. The radiation detection nozzle and the thermometer protection nozzle were also attached to the headers. **Figure 7** gives an exterior view of the mock-up of superheater outlet header and the longitudinal macrostructure of the circumferential welded joint of the central header. There were no welding defects such as cracks in the circumferential weldment with a thickness of 138 mm, indicating the soundness of welded joints. The mock-up fabrication confirmed the practical applicability of the developed welding and bending techniques.[7]

Figure 6 Bend processing of the HR6W large-diameter pipes

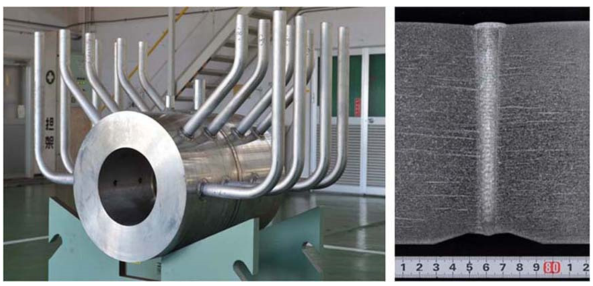

Figure 7 Exterior view of the HR6W superheater outlet header mock-up and the longitudinal cross-sectional macrostructure of the circumferential weldment

3.3 Evaluation technique of sensitivity to reheat cracking

In the field testing conducted in Europe ahead of Japan, there were cases in which cracks resulted in the welded joints of the Alloy 617 pipes.[8] Cracking is considered to occur when residual stress at the weld is relaxed during operation. Ni-based alloys are generally more vulnerable to solidification cracking or the aforementioned reheat cracking than ferritic steels, which are used in the major piping of conventional boilers. Despite the necessity of sufficient examinations on Ni-based alloys, there are no quantitative evaluation methods. We have therefore developed a method to evaluate the reheat cracking sensitivity of each Ni-based candidate alloy. To simulate welding residual stress, a test piece is strained by three-point bending and is heated to 700°C (which is the temperature of A-USC operation) while keeping the bent test piece as-is. In this way, the presence/absence of cracks can be examined regularly as the assessment of reheat cracking sensitivity.[9]

The initial strain corresponds to the magnitude of residual tension. If a small initial strain can yield a crack in a material and it happens within a short while, the material is considered highly sensitive to reheat cracking. The creep strain at the occurrence of cracks can be estimated by FEM analysis. Based on the results, a graph that shows the relationship between the initial strain and the creep strain can be obtained, thereby enabling the quantitative estimation of the risk of reheat cracking at the welds of actual units. With this method, it is also possible to evaluate the effect of suitable countermeasures (such as post-weld heat treatment).

3.4 Evaluation of long-term creep strength

Creep strength should be taken into consideration when designing A-USC boiler piping and headers. In so doing, it is important to examine the creep strength of welded joints, bends and welded metals, as well as base metals. Given below are the results of welded joint examinations.

(1) Plate welded joints

With respect to the welded joint of each test material, long-term creep testing is under way to evaluate the creep rupture strength at 100,000 hours, which is an indicator of allowable tension stress in boiler materials. As an example, **Figure 8** gives the 700°C creep rupture properties of plate welded joints of Ni-based candidate alloys and the cross-sectional macrostructure.[10] At 700°C, Alloy 263, Alloy 740 and Alloy 141 are especially superior in creep rupture strength, which are then followed by Alloy 617, HR35 and HR6W in sequence. When the temperature was further elevated, differences in the strength among the alloys tended to diminish. For Ni-based alloys, there have been no reports about creep strength reduction due to HAZ failure, whose occurrence was reported regarding the welds of high-strength ferritic heat-resistant steels. In several types of Ni-based alloys, however, HAZ ruptures occurred in the welds during long-term creep rupture testing, indicating the importance of verification of welded joint strength due to 100,000 hours of continuous examination.

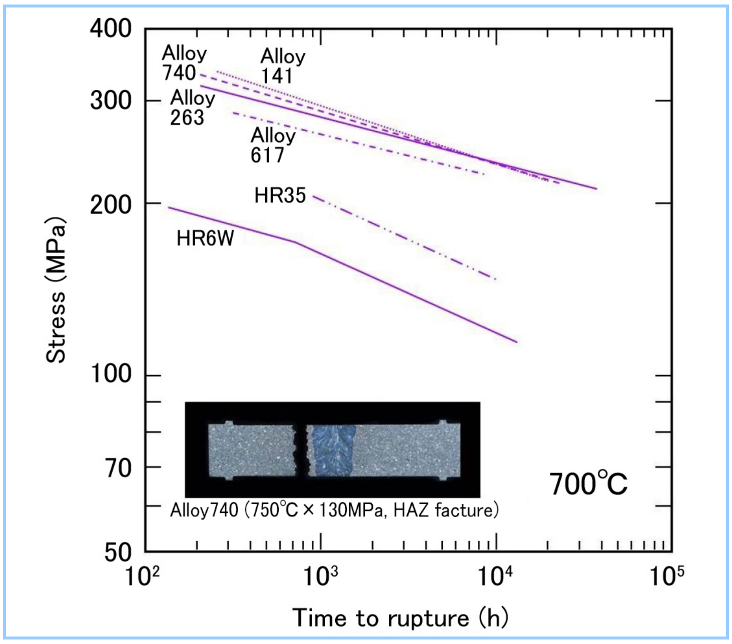

Figure 8 Creep rupture properties and the cross-sectional macrostructure of the Ni-based alloy plate welded joints

(2) Large-diameter pipe welded joints

Figure 9 compares the creep rupture properties of HR6W large-diameter pipe welded joints with that of the base metal. When also including the data on small-diameter tube joints (which was obtained on occasions other than the national project) in addition to Figure 9, the maximum assessment duration of creep rupture testing reaches 70,000 hours.[6] With HR6W, the creep strength of welded joints is as good as or superior to the base metal throughout the temperature range of 700°C to 800°C. According to the extrapolation results of creep rupture data, the 700°C creep rupture strength at 100,000 hours is expected to satisfy the target strength of 90 MPa or higher in the national project. Among advanced ferritic steels, SAVE12AD is the most promising, and the data on its creep rupture strength at approximately 30,000 hours has already been collected using the large-diameter pipe welded joints. The testing is still ongoing to obtain the results of 100,000 hours.

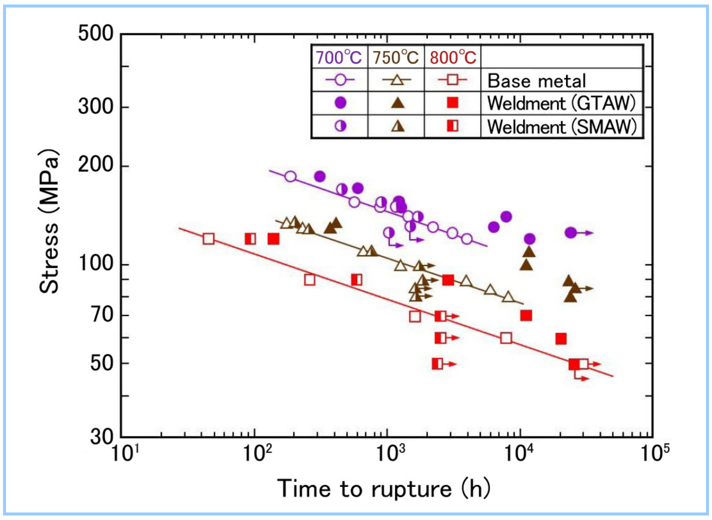

Figure 9 Creep rupture properties of the HR6W large-diameter pipe welded joints

3.5 Evaluation of steam oxidation and high-temperature corrosion properties

Steam oxidation scale is formed on the inner surface of high-temperature heat transfer tubes and piping as a result of the reaction with superheated steam. When heat transfer is prevented by scale formation, the metal temperature increases, accelerating the occurrence of creep damage. The deposition of exfoliated scale may result in blocked tubes/pipes or steam turbine erosion. When used in A-USC boilers, the Ni-based candidate alloys are supposed to be exposed to temperatures of 700°C or above, which is higher than conventional steel. As these alloys contain more added elements than the currently-used austenitic steels, many unclear points of their behavior including the growth rate and properties of scale are still unknown. We therefore examined the steam oxidation properties of Ni-based candidate alloys through laboratory testing in a high-temperature steam environment.

In testing, a test piece was placed in a reaction tube and was exposed to hot steam of a predetermined temperature. An electric furnace was used to produce hot steam by superheating ion-exchanged water in which the dissolved oxygen level is reduced by N_2 gas. The testing was conducted at three different temperatures (700°C, 750°C and 800°C) with a maximum test duration of 10,000 hours. After the test completion, the tendency of scale growth was measured as the average cross-sectional thickness of the inner scale layer, whereas scale properties were evaluated by composition analysis. As the steam oxidation properties of the test materials, the average thicknesses of steam oxidation scale (inner layers) after 10,000 hours of steam exposure are shown in **Figure 10**.[11] In the 700°C or 750°C steam environment, which is similar to that of A-USC boiler components subject to high temperatures, no significant differences were seen in the scale thickness of these Ni-based alloys (approximately 5 μm thick in general). When compared with the currently-used austenitic steel (25Cr steel or KA-SUS310J1TB), the Ni-based alloys yielded thinner scale and therefore exhibited superior steam oxidation resistance. Although Ni-based alloys (excluding HR6W) yielded slightly thicker scale than 25Cr steel at 800°C for 10,000 hours, the thicknesses of the formed layers were approximately 10 μm, which indicates that no practical problems will be caused.

Unlike piping, in the heat transfer tubes, which are directly exposed to the flame, the temperature of the outer surface is higher than the steam temperature inside. The heat transfer tubes of the final superheater or reheater are expected to be at a maximum temperature of 750°C to 800°C. Generally speaking, Ni-based alloys have excellent corrosion resistance, but at such high temperatures they are likely to be subjected to oxidation and high-temperature corrosion due to coal ash. Therefore, through hot corrosion testing while taking various coal fuel properties into consideration, the high-temperature corrosion properties were compared between the test materials and existing materials. The test pieces were coated with the prepared standard ash (1.5 mol Na_2SO_4 + 1.5 mol K_2SO_4 + 1.0 mol Fe_2O_3) in a gaseous atmosphere containing SO_2 of 0.20 vol%. The test temperatures (650°C and 700°C) were determined in accordance with the range in which coal ash will melt and cause especially severe corrosion. After the test was completed, the appearance and weight loss after the removal of scale were examined.

As the high-temperature corrosion properties of the test materials, the relationship between the Cr content and the average weight loss per a given unit in the corrosion test is shown in **Figure11**[12]. The Cr content is the primary factor to determine the corrosion resistance of each test material (Cr is a major constituent element), whereas weight loss indicates the corrosion wastage. As the Cr content increases, the corrosion wastage diminishes. The Ni-based alloys with a high Cr content exhibited good corrosion resistance. Unlike the results at 650°C, those of 700°C showed a tendency that when the Cr content was identical, corrosion wastage was slightly larger in Ni-based alloys than in austenitic steels. This is considered to result from the high cross-reactivity between Ni and S in SO_2 gas and the corrosion process facilitated by the melting of corrosion products. However, their corrosion resistance does not considerably exceed that of existing austenitic steels at 650°C (equivalent to the USC exposed environment). Therefore, no practical problems will be caused.

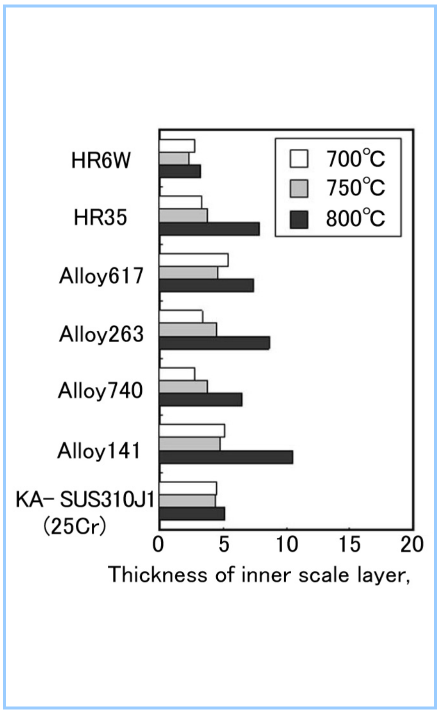

Figure 10 Steam oxidation property

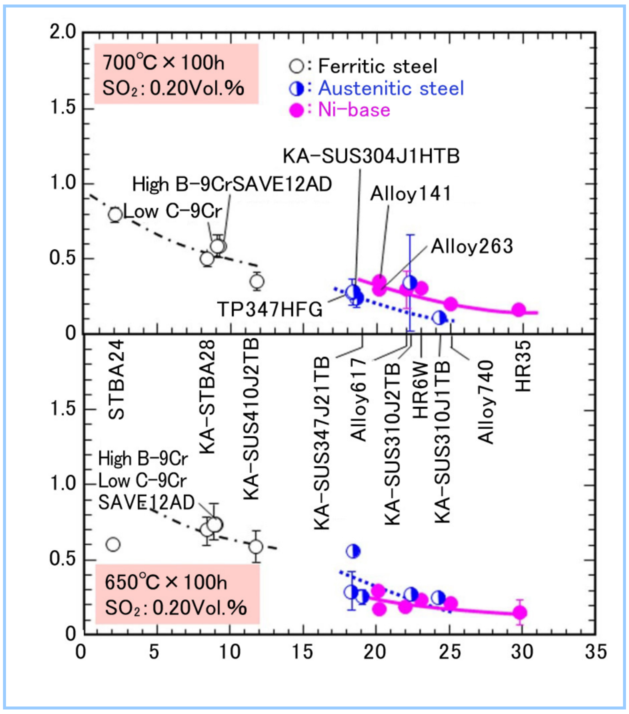

Figure 11 High-temperature corrosion property

4. Field testing

To verify the candidate materials and developed manufacturing techniques, we started field testing in May 2015 in cooperation with the Mikawa Power Station of the Sigma Power Ariake in Omuta City, Fukuoka, Japan. In this testing, heat transfer tube panels were attached to the actual boiler. Operation is under way in the 700°C steam environment involving other components such as the header, piping, high temperature valve and turbine casing. Inserted into the panels are the A-USC candidate materials (HR6W, HR35, Alloy 617, Alloy 263 and Alloy 740) and the currently-used austenitic steels (KA-SUS304J1HTB and KA-SUS310J1TB) for comparison. HR6W is used for the piping test, and the hot bending parts and circumferential welded joints manufactured by our company are also inserted as test materials. The other components included are HR6W high-temperature valves and Alloy 625 turbine casing.

Figure 12 is a photograph of the actual unit with the HR6W pipes inserted. The planned test duration is approximately 10,000 hours. After completion, the tubes and pipes will be removed for investigation. The data on corrosion resistance and material properties, which shows the material change with time, will be collected to verify the soundness of A-USC boiler materials.

Along with the field testing, we started developing technologies for creep damage assessment and non-destructive inspection, as maintenance technologies for Ni-based alloys (candidate materials for use in piping). In the creep damage assessment, the elementary process during the creep of Ni-based alloys and the fracture morphology are examined, and the applicability of existing life assessment methods such as the creep void method and the hardness method is also under examination using materials with creep interrupted. For the non-destructive inspection methodology, on the other hand, we are evaluating the applicability of ultrasonic waves to Ni-based alloys, through which ultrasonic waves is difficult to be transmitted. We will keep working on the development of these technologies to make them practically applicable. A new Ni-based alloy with a permissible upper temperature of 800°C (USC800), which exhibits both superior high strength and hot processability, is also under development to make it practically viable[13]. Such development of new materials will give us the capability to satisfy the need for the further elevation of the steam temperature.

Figure 12 Field testing with the inserted HR6W large-diameter pipes

5. Conclusion

To realize A-USC boilers, which is the key technology for the increased efficiency of coal-firing power generation, we have undertaken research for the practical application of boiler materials. The manufacturing techniques were established and the properties of long-term creep strength, high-temperature corrosion and steam oxidation were evaluated. The practical applicability of tested materials has been indicated. With respect to the long-term creep strength, testing will be continued to obtain the creep rupture strength at 100,000 hours. Field testing has been carried out since May 2015 to verify the soundness of A-USC boiler materials. We will further develop technologies for creep damage assessment and non-destructive inspection for maintenance purposes, aiming to realize the early commercial use of A-USC boilers.

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