Article:

Lee, T-H., Suh, H-Y., Han, S-K. et al. (2 more authors) (2016) Effect of a heat treatment on the precipitation behavior and tensile properties of alloy 690 steam generator tubes. Journal of Nuclear Materials, 479. pp. 85-92. ISSN 0022-3115

https://doi.org/10.1016/j.jnucmat.2016.06.038

Effect of a heat treatment on the precipitation behavior and tensile properties of alloy 690 steam generator tubes

Tae-Hyuk Lee[a,b], Ho-Young Suh[c], Seul-Ki Han[a,d], Jae-Soo Noh[e], Jong-Hyeon Lee [a,d*]

[a]*Department of Materials Science & Engineering, Chungnam National University, 99 Daehak-ro, Yuseong-gu, Daejeon, 34134, Republic of Korea*
[b]*Department of Materials Science & Engineering, Robert Hadfield Building, University of Sheffield S1 3JD, United Kingdom*
[c]*Division of Electron Microscopic Research, Korea Basic Science Institute, Daejeon, 34133, Republic of Korea*
[d]*Rapidly Solidified Materials Research Center Chungnam National University, Rapidly Solidified Materials Research Center, Daejeon, 34134, Republic of Korea*
[e]*New Project Center, KEPCO Nuclear Fuel Company Ltd., 989 Daedeokdae-ro, Yuseong-gu, Daejeon, 34039, Republic of Korea*

*Corresponding author: Jong-Hyeon Lee
Address: Chungnam National University, Daejeon, Republic of Korea
Phone: +82-42-821-6596
Fax: +82-42-822-5850
E-mail: jonglee@cnu.ac.kr

Abstract

The intergranular carbide precipitation behavior and its effect on the tensile properties were investigated in alloy 690. The precipitation of intergranular carbides, identified as Cr-rich $M_{23}C_6$, was retarded on the low-angle grain boundaries and the coincidence-site lattice boundaries. The $M_{23}C_6$ carbides have a cube-cube orientation relationship with the matrix. The ultimate tensile strength, yield strength, and elongation of the solution annealed alloy 690 are 648.2±8.2 MPa, 242.8±10.5 MPa and 44.9±2.3 %, respectively. The ultimate tensile strength and the yield strength increased to 764.8±7.8 MPa and 364.8±10.2 MPa until the aging time reached 16 hours. This increase is ascribed to the $M_{23}C_6$ carbide acting as reinforcements. However, when the aging time exceed 16 hours, these properties gradually decreased with increasing aging time. The decrease in ultimate tensile strength, yield strength, and elongation were mainly caused by the intergranular cracking due to the low bond strength between the carbide and the matrix.

Keywords: Alloy 690, heat treatment, $M_{23}C_6$ carbide, grain boundary character, tensile properties.

1. Introduction

Alloy 690, a nickel-based alloy having high chromium content (27-31 wt.%), was developed as a replacement for alloy 600 in the steam generators of pressurized water reactors (PWRs) in nuclear power plants, and is now widely used owing to its excellent resistance to stress corrosion cracking (SCC), pitting corrosion, and corrosion fatigue in aggressive primary water environments [1-3]. However, the mechanical properties of alloy 690 are noticeably inferior to those of alloy 600 [4] while its chemical properties are significantly improved. The mechanical properties of Ni-based alloys are dependent on the morphology and distribution of precipitates in the matrix [5]. Generally, under the conditions of heat treatment or during high-temperature service, $M_{23}C_6$, as a typical carbide commonly precipitates at the grain boundary in alloy 690 [6-8].

The precipitation of second phases, such as carbides of $M_{23}C_6$, is known to be generated selectively at grain boundaries during thermal aging [9]. It was found that morphology and the precipitation kinetics of $M_{23}C_6$ carbides are closely related to the grain boundary character and thermal aging temperature [8]. Hu et al. demonstrated that the coherent orientation relationship between $M_{23}C_6$ and the matrix plays an important role in the precipitation morphology of $M_{23}C_6$ in a Ni-Cr-W alloy aged at 700 °C [5]. Hong et al. found that the carbide morphology and size are strongly related with grain boundary misorientation in austenitic stainless steel [10]. Trillo and Murr found that carbides tend to precipitate first at random grain high-angle boundaries and then at non-coherent twin boundaries in 304 stainless steels aged at 670 °C [11]. To date, it is still unclear why the morphology of $M_{23}C_6$ is different at different types of grain boundaries in various alloys during thermal aging. Also, the precipitation behavior could be affected by the alloy composition (such as carbon contents) and heat treatment conditions. In this light, the precipitation behavior of $M_{23}C_6$ carbide warrants further investigation.

Several studies on alloy 690 have been carried out with most of them focusing on aspects related to the influence of $M_{23}C_6$ carbides on corrosion properties [12], fretting wear [13] and fatigue crack growth [14]. The role of the different morphologies of the $M_{23}C_6$ of alloy 690 is very complex. Hence, it is important to study the $M_{23}C_6$ carbide precipitation behavior during thermal aging and its effects on the mechanical properties of alloy 690 for application to steam generator tubes.

In this study, we have investigated the time-temperature-precipitation (TTP) curve of alloy 690 tubes for carbon content of 0.013 wt.%. The influence of the grain boundary character on $M_{23}C_6$ precipitation behaviors in an alloy 690 was then studied by the electron backscatter diffraction (EBSD) technique. Finally, we have determined the effects of the intergranular carbide behaviors on the tensile properties of alloy 690 tubes.

2. Experimental

The material used in this work was commercial alloy 690 tubing (o.d.: 19 mm and i.d.: 16 mm) provided by KEPCO Nuclear Fuel Company in South Korea. The chemical composition of the alloy 690 is listed in Table 1. The specimens were solution annealed at 1100 °C for 30 min in an argon atmosphere followed by water quenching. After this, the specimens were aged in an argon atmosphere at various temperatures from 500 °C to 1000 °C for various periods from 1 to 50 hours followed by water quenching. Prior to the scanning electron microscopy and the EBSD experiments, the specimen surface was mechanically polished using #1200 emery papers, and then electro-polished in an electrolyte (100 mL distilled water + 25 mL H_2SO_4) for 30-40 s at 6 V with a platinum plate cathode. During the electro-polishing process, the temperature of the electrolyte was maintained at 20 °C by using a cooling system. The specimen was then ultrasonically cleaned in acetone and ethanol for 10 minutes in order to remove small particles on the surface.

The characterization of grain boundary misorientation and the orientation relationships of the carbides was carried out with a field emission scanning electron microscope (FE-SEM, JEOL, JSM 7000F, Japan) utilizing an EBSD (INCA, U.K.) technique. The coincidence-site lattice (CSL) boundaries were then classified using Brandon`s criterion; ∑3 to ∑49 were considered in this study [15-17]. The structure and morphology of the precipitated carbides were analyzed by transmission electron microscopy (TEM, FEI, Tecnai G2 F30, U.S.A). Further, the distribution of phases and grain orientation maps were obtained with an automatic crystal orientation mapping (ACOM, Nanomegas, ASTAR, Belgium) system installed in a TEM. The TEM sample was fabricated by an analytical dual beam focused ion beam scanning electron microscope (FIB/SEM, Tescan, LYRA3XMU, Czech) system typically 10 μm long and 2 μm wide. The bulk-alloy 690 (thickness: 1 mm) was oriented with the basal planes parallel to the sample holder surface. The sample was milled with a Ga ion beam at an accelerating voltage of 30 keV. During the sample preparation steps, care was taken to minimize gallium damage to the sample surface by positioning a platinum layer over the region of interest. Finally, the TEM sample was lifted out in situ and placed on a TEM grid. Furthermore, tensile properties were measured using a universal testing machine at room temperature. The measured engineering stress and strain data were automatically recorded by a computer. The tensile test was conducted in displacement control with a strain rate of 1x 10^{-3} S^{-1}.

3. Results & Discussion

Fig. 1 shows the time-temperature-precipitation (TTP) curve of alloy 690 tubes with a carbon content of 0.013 wt.%. The approximate time necessary for the carbides to precipitate and then become nearly semi-continuous along the grain boundary was 30 hours at 650 °C, 1 hour at 750 °C, 5 hours at 850 °C and 45 hours at 925 °C. It should be noted that no intergranular carbide was generated on the grain boundaries in the alloy 690 aged at temperatures of 500-600 °C under our experimental conditions. Frequently, the published isothermal TTP diagrams show a "C-curve", as shown in Fig. 1. This is a consequence of the interaction between diffusion coefficient of alloying elements and the number of nucleation sites. At low temperatures, diffusion coefficient of elements is low and the number of nucleation sites of precipitates is high. On the contrary, at high temperature, diffusion coefficient of alloying elements is high and the number of nucleation sites is low. Optimal conditions for precipitation processes are located at intermediate temperatures [18].

Generally, the precipitation behavior is strongly dependent on both the service temperature and the alloying elements [19]. The alloying elements Cr, Mo and Ti all form carbides with substantially higher enthalpies than iron carbides, while the elements Ni, Co and Cu do not form carbide phases [20], as metallic alloying elements cannot diffuse sufficiently rapidly enough to allow alloy carbides to nucleate in alloy 690 tubes aged at temperature of 500-600 °C in our experimental periods. A substitutional-diffusion controlled reaction is necessary for the formation of the alloyed $M_{23}C_6$ carbide. This reaction occurs mostly at high temperatures and requires a sufficiently long time at low temperatures. Thus, the $M_{23}C_6$ carbide can be considered as a metastable phase at lower temperatures. Therefore, high temperatures and a long enough time are needed for the diffusion of the alloy elements prior to the nucleation growth of the alloy carbides. As a result, the approximate time required for the carbides to precipitate was decreased with an increase in the aging temperature. However, as the aging temperature increased further to 750 °C, the approximate time was increased with an increase in the aging temperature. When the aging temperature increased, the number of nucleation sites of $M_{23}C_6$ carbides was decreased. The solid solubility of carbon in the Ni-based matrix was increased as the temperature was increased [21]. Thus, the amount of carbon precipitates decreased as the aging temperature increased. As a consequence, the nucleation and subsequent growth of the $M_{23}C_6$ carbides becomes difficult at high temperature. Therefore, the approximate time for the carbides to precipitate was increased with an increase in the aging temperature.

Fig. 2 shows the microstructure and carbide precipitation behavior at the grain boundaries in alloy 690 after an aging treatment at 700 °C for various periods ranging from 1 to 50 hours. After an aging treatment for 1 hour, no carbide was found on the grain boundaries under the experimental conditions used here, as shown in Fig. 2(a). As the aging time increased to 5 hours, tiny and semi-continuous carbides precipitated along the grain boundaries, as shown in Fig. 2(b). When the aging time was increased to 10 hours, the carbides that precipitated along the grain boundary became coarse, as shown in Fig. 2(c). When the aging time reached 20 hours, some carbides grew into the matrix near one or both sides of the grain boundaries, as shown in Fig. 2(d). When the aging time reached 50 hours, it was noted that the carbides grew into the matrix mainly on the grain boundaries of alloy 690, as shown in Fig. 2(e).

Fig. 3 shows the average grain size excluding the twins of alloy 690 tubes after an aging treatment at 700 °C for various periods ranging from 1 to 50 hours. The average grain size of the solution annealed specimen was 33.2 ± 6.2 µm. However, there was no additional grain growth during the aging treatment with an increase in the aging time. With an increase in the aging time, the average grain size increased slightly to 39.4 ± 7.4 µm, within the error range. This result is due to the effect of the intergranular carbides. Intergranular carbides are likely quite effective in pinning the boundaries against migration. This may explain why the average grain size did not increase after the aging treatment at 700 °C for 50 hours. Moreover, it may explain why the aging-treated specimens had significant inhomogeneity in the grain size, and why some grains were abnormally large, as shown in Fig. 2.

Fig. 4 shows a bright-field TEM image, the selected area electron diffraction (SAED) pattern and the energy-dispersive X-ray spectroscopy (EDS) mapping results of the intergranular $M_{23}C_6$ carbide in the alloy 690 aged at 700 °C for 5 hours. Fig. 4(b) is taken from the circled area in Fig. 4(a). From the SAED pattern analysis and EDS results, the intergranular carbide was identified as chromium rich (Cr-rich) $M_{23}C_6$ carbide. $Cr_{23}C_6$ has the same face-centered cubic structure as that of the alloy 690 matrix, with a lattice constant of 1.056 nm [22], which is approximately three times larger than that of a matrix of 0.353 nm [23]. The Cr-rich $M_{23}C_6$ has a cube-cube orientation relationship of $<111>_{M23C6}$ // $<111>_{matrix}$, $\{111\}_{M23C6}$ // $\{111\}_{matrix}$ with one of the neighboring grains.

Fig. 5 indicates the orientation relationship between a carbide and its neighboring grains in alloy 690 aged at 700 °C for 5 hours. The orientation relationship between a carbide and its neighboring grains can easily obtained be from an ACOM analysis. It clearly shows that the carbide and grain 1 are coherent, whereas the results show incoherency with grain 2 and grain 3. According to the above analysis, when $M_{23}C_6$ carbide nucleates on the grain boundaries and has a $\{111\}_{M23C6}$ // $\{111\}_{matrix}$ coherent orientation relationship with the matrix of a coherent grain, it grows into the matrix of an incoherent grain. Cr atoms can easily substitute for Ni and Fe atoms at the incoherent grain interface and promote the growth of $M_{23}C_6$ carbide, due to the assistant flux of vacancies and the high interface energy at the incoherent phase interface [24]. For this reason, the cellular carbide that grows into one of the two grains forms on the grain boundaries in alloy 690, as shown in Fig. 2(d,e).

Fig. 6 shows a SEM image of the morphology, indicating that the size of the carbides was distinctly related to the grain boundary character in alloy 690 aged at 700 °C for various periods. It is clear that the intergranular $M_{23}C_6$ carbides are closely related to the grain boundary character. Furthermore, the figure shows that the high-angle boundaries with a misorientation angle of 15° to 60° serve as preferential sites for intergranular $M_{23}C_6$ carbides apart from the CSL boundaries. Furthermore, the low-angle boundaries are immune to intergranular carbides before the aging time reaches 5 hours. In terms of the CSL boundaries, carbides were not found on the ∑3 boundaries under the experimental conditions. On the other hand, tiny carbides precipitated on the ∑9 and ∑27 boundaries. However, the carbide size on the CSL boundaries was smaller than that on the random high-angle boundaries.

Generally, the precipitation behaviors at grain boundaries are believed to be related to the atomic arrangement between the neighboring grains and the grain boundary energy. A relatively high degree of the atomic matching of the low-angle boundaries (<15°) and the CSL boundaries would lead to lower grain boundary energy in comparison with the high angle boundaries apart from CSL boundaries [20]. Furthermore, it is well known that all atoms at the ∑3 boundary fit perfectly into both grains, resulting in extremely low boundary energy and low mobility [22]. The grain boundary energy then provides the requisite energy for the nucleation growth of the intergranular $M_{23}C_6$ carbide. Moreover, the growth of intergranular $M_{23}C_6$ carbides along the grain boundaries may be controlled by the activation energy of carbide nucleation at grain boundaries with different characteristics. As a result, the precipitation of $M_{23}C_6$ carbides is retarded on the low-angle boundaries and the CSL boundaries, unlike that on the high-angle boundaries. As the degree of grain boundary misorientation increases, the constraints on the intergranular carbides are reduced due to the larger grain boundary energy. Therefore, the intergranular $M_{23}C_6$ carbides are closely related to the grain boundary characteristics.

The ultimate tensile strength, yield strength, and elongation outcomes of the alloy 690 aging-treated at 700 °C with various aging times are shown in Fig. 7. As shown in Fig. 3, the average grain size did not increase with an increase in the aging treatment time. Therefore, in the present work, the effect of the grain size on the tensile properties of alloy 690 was considered negligible. The ultimate tensile strength, yield strength and elongation of the solution annealed alloy 690 are 648.2±8.2 MPa, 242.8±10.5 MPa and 44.9±2.3 %, respectively. After an aging treatment, the ultimate tensile strength and the yield strength of alloy 690 increased to 764.8±7.8 MPa and 364.8±10.2 MPa, respectively, until the aging time reached 16 hours. However, when the aging time exceeded 16 hours, the ultimate tensile strength and yield strength gradually decreased to 731.5±12.5 MPa and 335.6±8.4 MPa, respectively. On the other hand, the elongation consistent decreased to 30.4±2.1 % with an increase in the aging time, as shown in Fig. 7(b).

The above results indicated that the variation of the tensile strength with the aging time is related to the morphology and amount of $M_{23}C_6$ carbides. If the precipitate carbides are smaller and they exist in greater amounts, the precipitation strengthening effect will be more significant [25]. During the aging treatment (1-16 hours), the precipitation of the discontinuous $M_{23}C_6$ carbides on the grain boundaries blocked the dislocation motion to produce an obvious pinning strengthening effect [26]. For this reason, the ultimate tensile strength and yield strength increased with the aging time when it reached 16 hours. Nevertheless, the ultimate tensile strength and the yield strength decreased when the aging time exceeded 16 hours. When the coarsened and cellular carbides precipitate at the grain boundaries, many chromium and carbon elements in the matrix are consumed and the chromium and carbon concentration decreases. As a result, the crystal lattice distortion of the matrix is reduced. Chromium and carbon play important roles in the strength of alloys, and the strength decreases with a decrease in the chromium and carbon concentration in the alloys [27]. Moreover, it is well known that the coarsened and cellular carbide is brittle and that there is low bond strength between the carbide and the matrix [5].

Fig. 8 shows the microstructure of the tensile test specimen near the fracture. A number of micro-cracks are generated at the grain boundaries. Typically, the matrix grains are connected through grain boundaries before an aging treatment. However, the grains are separated by the coarsened intergranular carbide after the aging treatment due to the brittle carbides and low bonding strength between the coarsened carbide and the matrix. When the aging time reaches 20 hours, the coarsened carbides precipitate along the grain boundaries, as shown in Fig. 6. During the tensile test, when the tensile stress is sufficiently high, cracks are generated at the interface between the coarsened carbide and the matrix, as shown in Fig. 8(e). Therefore, the ultimate tensile strength and yield strength decreased when the aging time exceeded 16 hours due to the generation of the coarsened carbide at the grain boundaries, as shown in Fig. 7(a).

Fig. 9 shows the fracture surfaces of the tensile test specimens prepared with different aging times after tensile testing to failure at room temperature. After an aging treatment for 1 hour, the fracture surface exhibits a finely dimpled surface, which is a typical characteristic of the dimpled ductile mode of failure associated with good plastic deformation, as shown in Fig. 9(a). However, when the aging time was increased to 5 hours, failure occurred in a mixed intergranular and transgranular mode due to the precipitation of intergranular carbides. In Figs 9(b-f), it is clear that the intergranular fracture areas on the fracture surface increased with an increase in the aging time because the intergranular $M_{23}C_6$ carbide precipitated and coarsened at the grain boundary. During the tensile test, the stress concentration at the grain boundaries formed by accumulated stresses at the grain boundary slip band intersection will lead to separation between the carbide and the matrix (Fig. 8(e)) [28]. It can also be observed seen that dimple structures predominate on the fracture surface of the matrix, as shown in Figs. 9(e, f). This can be attributed to the decreased chromium and carbon concentration in the matrix due to the coarsened intergranular $M_{23}C_6$ carbides at the grain boundary during the aging treatment. In addition, the precipitation of coarsened and cellular $M_{23}C_6$ carbides retarding the occurrence of necking is detrimental to the ductility of the alloy 690. Thus, the ductility of alloy 690 tubes is closely dependent on the intergranular cracking associated with cavitation at the interface between the dense $M_{23}C_6$ carbide and nickel-based matrix. Hence, the elongation decreased with an increase in the aging time, as shown in Fig. 7(b).

4. Conclusion

In this study, we investigated the time-temperature-precipitation (TTP) curve of the $M_{23}C_6$ carbides and related effects on the tensile properties in alloy 690 tubes with a carbon contents of 0.013 wt.% by an aging treatment in a temperature range from 500 °C to 950 °C for various periods ranging from 1 to 50 hours. The results are as follows:

1) In an aging temperature range of 650-925 °C, most of the intergranular carbides generated on the grain boundaries, while no precipitated carbide was observed under 600 °C and over 925 °C. The carbides precipitated on the grain boundaries were identified as chromium-rich $M_{23}C_6$ carbides.

2) The intergranular $M_{23}C_6$ carbides tended to form preferentially at high-angle boundaries apart from the CSL boundaries while they were absent at the twin boundaries under our experimental conditions.

3) The intergranular carbides function as reinforcement and provide strength enhancements when the aging time reached 16 hours. As the aging time exceeded 16 hours, however, the ultimate tensile strength and the yield strength gradually decreased with an increase in the aging time.

4) After aging treatment, the elongation of alloy 690 decreased with increase in the aging time. The decrease in the ultimate tensile strength, yield strength and the elongation were mainly caused by the intergranular cracking due to the low bond strength between the coarsened intergranular carbide and the matrix.

Acknowledgments

[†]This work was supported by the research fund of 2015 Chungnam National University in

South Korea.

REFERENCES

[1] D.L. Harrod, R.E. Gold, R.J. Jacko, Alloy optimization for PWR steam generator heat-transfer tubing, JOM, 53 (2001) 14-17.

[2] J.J. Kai, G.P. Yu, C.H. Tsai, M.N. Liu, S.C. Yao, The effect of heat treatment on the chromium depletion, precipitate evolution, and corrosion resistance of INCONEL alloy 690, Metall. Trans. A, 20 (1989) 2057-2067.

[3] P.I. Andersen, M.M. Morra, Stress corrosion cracking of stainless steels and nickel alloys in high-temperature water, Corrosion, 64 (2008) 15-29.

[4] P.H. Berge, J.R. Donati, Materials requirements for pressurized water reactor steam generator tubing, Nucl. Technol. 55 (1981) 88-104.

[5] R. Hu, G.H. Bai, J.S. Li, J.Q. Zhang, T.B. Zhang, H.Z. Fu, Precipitation behavior of grain boundary $M_{23}C_6$ and its effect on tensile properties of Ni-Cr-W based superalloy, Mater. Sci. Eng. A, 548, 83 (2012).

[6] Y.S. Kim, J.S. Kim, H.P. Kim, H.D. Cho, The effect of grain boundary misorientation on the intergranular $M_{23}C_6$ carbide precipitation in thermally treated Alloy 690, J. Nucl. Mater. 335 (2004) 108-114.

[7] T.M. Angeliu, G.S. Was, Behavior of grain boundary chemistry and precipitates upon thermal treatment of controlled purity alloy 690, Metall. Trans. A, 21 (1990) 2907-2107.

[8] H. Li, S. Xia, B.X. Zhou, W.Q. Liu, C-Cr segregation at grain boundary before the carbide nucleation in alloy 690, Mater. Charact. 66, (2012) 68-74.

[9] X. Dong, X. Zhang, K. Du, Y. Zhou, T. Jin, H. Ye, Microstructure of carbides at grain boundaries in nickel based superalloys, J. Mater. Sci. Technol. 28, (2012) 1031-1038.

[10] H.U. Hong, B.S. Rho, and S.W. Nam, Correlation of M23C6 precipitation morphology with grain boundary characteristics in austenitic stainless steel, Mater. Sci. Eng. A. 318

(2001) 285–292.

[11] E.A. Trillo, L.E. Murr, A TEM investigation of $M_{23}C_6$ carbide precipitation behavior on varying grain boundary misorientation in 304 stainless steels, J. Mater. Sci. 33 (1998) 1263-1271.

[12] M. Casales, V.M. Salinas-Bravo, A. Martinez-Villafane, J.G. Gonzalez-Rodriguez, Effect of heat treatment on the stress corrosion cracking of alloy 690, Mater. Sci. Eng A. 332 (2002) 223-230.

[13] J.K. Hong, I.S. Kim, C.Y. Park, E.S. Kim, Microstructural effects on the fretting wear of Inconel 690 steam generator tube, Wear. 259 (2005) 349-355.

[14] H.B. Park, Y.H. Kim, B.W. Lee, K.S. Rheem, Effect of heat treatment on fatigue crack growth rate of Inconel 690 and Inconel 600, J. Nucl. Mater. 231 (1996) 204-212.

[15] D.G. Brandon, The structure of high-angle grain boundaries, Acta Metall. 14 (1966) 1479-1484.

[16] H. Grimmer, W. Bollmann, D.H. Warrington, Coincidence-site lattices and complete pattern-shift in cubic crystals, Acta Crystalogr. A. 30 (1974) 197-207.

[17] D.A. Smith, R.C. Pond, Bollmann`s O-lattice theory; a geometrical approach to interface structure, Int. Mater. Rev. 21 (1976) 61-74.

[18] B. Milkereit, L. Giersberg, O. Kessler, C. Schick, Isothermal Time-Temperature-Precipitation diagram for an aluminum alloy 6005A by in situ DSC experiments, Materials, 7 (2014) 2631-2649.

[19] J. Janovec, M. Svoboda, A. Vyrostkova, A. Kroupa, Time-temperature-precipitation diagrams of carbide evolution in low alloy steels, Mater. Sci. Eng. A. 420 (2005) 288-293.

[20] R.W. Balluffi, J.W. Cahn, Mechanism for diffusion induced grain boundary migration, Acta Metall. 29 (1981) 493-500.

[21] T.Y. Velikanova, A.A. Bondar, A.V. Grytsiv, The chromium-nickel-carbon (Cr-Ni-C)

phase diagram, J. Phase Equilib., 20 (1999) 125-147.

[22] A.L. Bowman, G.P. Arnold, E.K. Storms, N.G. Nereson, The crystal structure of $Cr_{23}C_6$, Acta Crystallographica B, 28 (1972) 3102-3103.

[23] Y. B. Lee, J.S. Jang, D.H. Lee, D.Y. Lee, I.H. Kuk, Selective precipitation of carbides in Alloy 690, J. Kor. Inst. Met. Mater., 35 (1997) 935-941.

[24] H. Li, S. Xia, B.X. Zhou, J.C. Peng, The growth mechanism of grain boundary carbide in Alloy 690, Mater. Charct. 81 (2013) 1-6.

[25] K. Maruyama, K. Sawada, J. Koike, Strengthening mechanisms of creep resistant tempered martensitic steel, ISIJ Int., 41 (2001) 641-653.

[26] J. Jiang, L.H. Zhu, Strengthening mechanisms of precipitates in S30432 heat-resistant steel during short-term aging, Mater. Sci. Eng. A, 539 (2012) 170-176.

[27] A.K. Jena, M.C. Chaturvedi, The role of alloying elements in the design of nickel-based superalloys, J. Mater. Sci. 19 (1984) 3121-3139.

[28] G. Terlinde, G. Luetjering, Influence of grain size and age-hardening on dislocation pile-ups and tensile fracture for a Ti-Al alloy, Metall. Mater. Trans. A, 13 (1982) 1283-1292.

Captions

Fig. 1. Time-temperature-precipitation curve of alloy 690 tubes for carbon content of 0.013wt.%

Fig. 2. Microstructure and precipitation behavior of alloy 690 tubes aged at 700 °C: a) 1 hour; b) 5 hours; c) 10 hours; d) 20 hours; e) 50 hours.

Fig. 3. Average grain size (excluding twins) of alloy 690 tubes according to aging time at 700 °C.

Fig. 4. TEM analysis of $M_{23}C_6$ carbide in alloy 690 tube aged at 700 °C for 5 hours: a) bright-field TEM image; b) SAED pattern; c) EDS mapping results.

Fig. 5. Orientation relationship between a carbide and its neighboring grains in alloy 690 aged at 700 °C for 5 hours: a) bright field TEM image; b) crystal orientation mapping image.

Fig. 6. Microstructure of intergranular $M_{23}C_6$ carbide behavior for different grain boundary characters in alloy 690 tubes aged at 700 °C for various aging periods.

Fig. 7. Tensile properties of alloy 690 tubes aged at 700 °C for various aging periods: a) ultimate tensile strength and yield strength; b) elongation.

Fig. 8. SEM micrograph of a region near the fracture of a tested specimen aged at 700 °C: a) 10 hours; b) 20 hours; c) 40 hours; d) 50 hours (white arrows indicated the cracks); e) high resolution image of intergranular cracks between the carbide and the matrix.

Fig. 9. Microstructure of fracture surfaces of tested specimens aged at 700 °C: a) 1 hour; b) 5 hours; c) 10 hours; d) 20 hours; e) 40 hours; f) 50 hours.

Table 1. The chemical composition of the alloy 690 tubes used in this experiment (wt.%).