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The Effects of Heat Treatments on the $M_{23}C_6$ Carbide Evolution and Grain Boundary Serration in Alloy 690

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1. Introduction

The predominant failure mode of the structural components in a nuclear power plant has been well known to be intergranular degradation such as intergranular stress corrosion cracking (IGSCC). Solid-solution strengthened nickel-based Alloy 690 (Ni–30wt%Cr–10wt%Fe) has become a substitute for Alloy 600 (Ni–16wt%Cr–8wt%Fe) as steam generator tubes and penetration nozzles owing to its excellent mechanical properties and corrosion resistance. Some laboratory tests revealed that Alloy 690 is resistant to IGSCC in various environments [1]. With a prolonged service life and improved performance being demanded by the nuclear energy industry, however, the need to improve the resistance to intergranular failure in Alloy 690 should also be considered. The present work is an attempt to elucidate the effects of various heat treatments on the evolutions of intergranular carbide precipitation and the grain boundary serration (GBS) in Alloy 690 to acquire a high resistance to intergranular degradations

2. Methods and Results

2.1 Experimental Procedures

Two types of Alloy 690 tubes (690-2C and 690-3C) with 19.05 mm in diameter and 1.08 mm thick were obtained in a 74.2 % cold-pilgered condition. Before heat treatment for carbide precipitation and GBS, all of the pre-existing Cr carbides were dissolved at high temperatures above the solution annealing temperature. From a previous study, the solution annealing (or Cr carbide dissolution) temperatures of these alloys were measured as 1055-1057 ℃ for 690-2C and 1106-1108 ℃ for 690-3C. Heat treatments were conducted in a box furnace equipped with a programmable controller, and two different courses of heat treatment were applied, that is, isothermal treatment and controlled cooling after solution annealing. The main objective of the isothermal treatment was to chronologically evolve Cr carbides depending on the grain boundary character. That of the controlled cooling treatment was to demonstrate the conditions for the occurrence of GBS and the effects of the carbon content, average grain size, and the cooling rate on the degree of GBS.

2.2 Microstructural features of Alloy 690

The results of the EBSD analysis on Alloy 690 are shown in Fig. 1. The types of grain boundaries were distinguished according to the misorientation angle between adjacent grains. Fig. 1(a) is an OIM image, which shows each grain in a different color. The grain boundaries with different CSL orientations are distinguished by colored lines in Fig. 1(b). The relative population of the LABs to the total of the boundaries was measured to be about 3.4 %, and that of the coherent TBs with a Σ3 CSL relationship (denoted as yellow lines in Fig. 1(b)) was 55.3 %. The rest of the boundaries were RHABs. In Alloy 690, most of the CSL boundaries were TBs, and many annealing twins were present owing to the considerably low stacking fault energy of this alloy [2]. Some twin-related boundaries such as Σ9 and Σ27 were also frequently observed in a specimen.

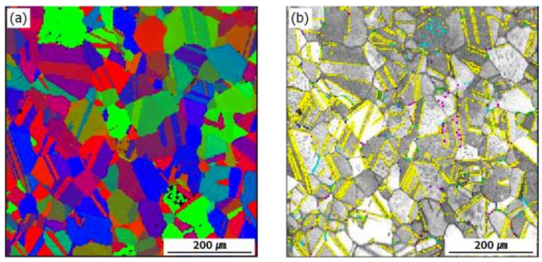

Fig. 1 EBSD results showing the grain boundary characteristics of 690-2C: (a) orientation image map and (b) grain boundary map denoted by the CSL model

2.3 $M_{23}C_6$ precipitation by isothermal treatments

Precipitation behaviors of Cr carbides during isothermal treatment at 720 ℃ were examined using 690-2C specimens. Precipitation morphologies depending on the aging time on the RHABs are shown in Fig. 4. Just after heat treatment of 0.1 hr, tiny and faceted carbides were found on the RHAB, and therefore, the carbide morphology corresponds to the 'early stage of growth' after nucleation. The carbides grow separately until they meet (Fig. 4(b)), which means that it is the 'stage of individual growth'. In Fig. 4(c), coarse and irregular carbides were semi-continuously distributed on the grain boundaries. Some of the faceted carbides appeared to be combined with each other, to form an irregular shape. Therefore, the carbide precipitation corresponds to the 'stage of concurrent coalescence and growth'. With an increased aging time, the combined carbides grew into a bulky and round shape (Fig. 4(d)), which means that it is the 'final stage of significant growth'.

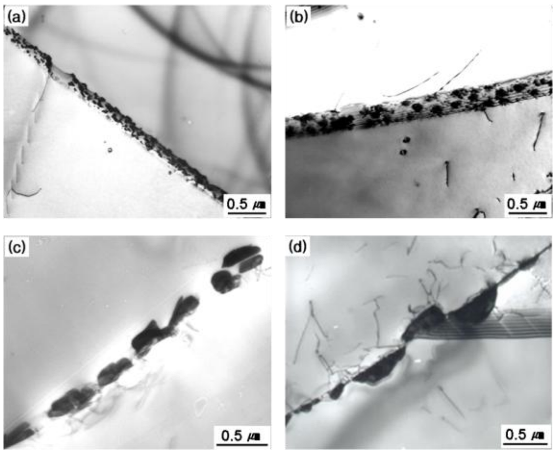

Fig. 4 Carbide precipitation on random high-angle grain boundaries in 690-2C after thermal treatments for (a) 0.1 hr, (b) 1 hr, (c) 10 hr, and (d) 100 hr at 720 ℃

2.3 3 GBS by controlled cooling treatments

Fig. 9 shows when and how GBS develops during the cooling process in this alloy. At a temperature of 1000 ℃ (Fig. 9(a)) after solution annealing, which was 106-108 ℃ below the solution annealing temperature of this alloy, the grain boundaries maintained a flat form without any Cr carbide precipitation along them. When the temperature reached 990 ℃ (Fig. 9(b)), some Cr carbides were initiated and subsequently grew within small fractions of some specific grain boundaries, and the grain boundaries began to be wavy, forming a serrated configuration owing to the carbide precipitation. As the temperature decreased, the fraction of the grain boundary being serrated increased with a concurrent growth of Cr carbides. At 900 ℃ (Fig. 9(c)), GBS expanded to most of the grain boundaries, except for CSL boundaries such as the coherent TBs, with a significant planar growth of Cr carbides. However, as the temperature decreased further to 800 ℃ (Fig. 9(d)), the degree of GBS remained unchanged. Therefore, it can be concluded that GBS occurs in a limited temperature range of 990-900 ℃ under the present conditions. It is known that the carbide precipitation on the serrated grain boundaries also depends on the grain boundary characteristic in Ni-based alloy [3].

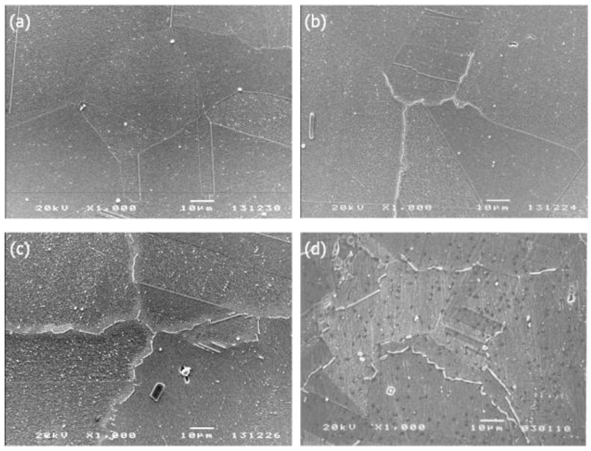

Fig. 9 Grain boundary morphologies and intergranular Cr carbide precipitations in Alloy 690-3C attained at (a) 1000 ℃, (b) 990 ℃, (c) 900 ℃, and (d) 800 ℃ during controlled cooling with a cooling rate of 0.5 ℃/min

3. Conclusions

By isothermal treatments at 720 ℃ for 0.1-100 hr after solution annealing, most of the grain boundaries except for the coherent twin boundaries were decorated with well-developed $M_{23}C_6$ carbides. The discontinuous precipitates were initiated on the grain boundary even by a heat treatment of 0.1 hr, and covered the entire grain boundary region within a heat treatment of 10 hr. With a long aging time of 100 hr, intragranular Cr carbides were precipitated on the imperfections such as the dislocations and stacking faults

GBS could be introduced by a slow cooling process in this alloy, and occurred in a limited temperature range of 990-900 ℃ under the present heat treatment conditions. The grain boundaries had a convex shape into the incoherent grain, from which it is believed that the grain boundary shape is closely associated with the grain boundary migration during serration in this alloy.

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