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Investigation of thermal aging effects on the tensile properties of Alloy 617 by *in-situ* synchrotron wide-angle X-ray scattering

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A B S T R A C T

The nickel-base Alloy 617 has been considered as the lead candidate structural material for the intermediate heat exchanger (IHX) of the Very-High-Temperature Reactor (VHTR). In order to assess the long-term performance of Alloy 617, thermal aging experiments up to 10,000 h in duration were performed at 1000 °C. Subsequently, *in-situ* synchrotron wide-angle X-ray scattering (WAXS) tensile tests were carried out at ambient temperature. $M_{23}C_6$ carbides were identified as the primary precipitates, while a smaller amount of M_6C was also observed. The aging effects were quantified in several aspects: (1) macroscopic tensile properties, (2) volume fraction of the $M_{23}C_6$ phase, (3) the lattice strain evolution of both the matrix and the $M_{23}C_6$ precipitates, and (4) the dislocation density evolution during plastic deformation. The property–microstructure relationship is described with a focus on the evolution of the $M_{23}C_6$ phase. For aging up to 3000 h, the yield strength (YS) and ultimate tensile strength (UTS) showed little variation, with average values being 454 MPa and 787 MPa, respectively. At 10,000 h, the YS and UTS reduced to 380 MPa and 720 MPa, respectively. The reduction in YS and UTS is mainly due to the coarsening of the $M_{23}C_6$ precipitates. After long term aging, the volume fraction of the $M_{23}C_6$ phase reached a plateau and its maximum internal stress was reduced, implying that under large internal stresses the carbides were more susceptible to fracture or decohesion from the matrix. Finally, the calculated dislocation densities were in good agreement with transmission electron microscopy (TEM) measurements. The square roots of the dislocation densities and the true stresses displayed typical linear behavior and no significant change was observed in the alloys in different aging conditions.

1. Introduction

The Very-High-Temperature Reactor (VHTR) is a leading candidate of the Gen-IV advanced reactors as proposed by the Next Generation Nuclear Plant (NGNP) program [1,2]. The VHTR is designed as a high-efficiency system with capabilities of providing process heat and coupling to hydrogen production facilities. The net plant efficiency of the VHTR can exceed 50% at 1000 °C, much greater than the 33% efficiency of current light water reactors. However, the high temperature environment poses challenges for reactor structural materials, especially when combined with high pressures and an aggressive atmosphere [3]. In particular, since the design lifetime of the VHTR is 60 years, high-temperature and long-term thermal-aging can induce significant degradation in the mechanical properties of the structural materials, and may pose potential challenges for long-term reactor operation.

One of the key components in the VHTR system is the intermediate heat exchanger (IHX), which is responsible for the heat transfer from the primary system to secondary systems [4,5]. The IHX is designed to function in an impure helium environment at temperatures up to 950 °C, and pressures around 7 MPa. Recently, Alloy 617, a solid-solution strengthened, nickel–chromium–cobalt–molybdenum alloy, has been selected as the structural materials for the IHX [6]. This alloy was developed in the 1970s for high-temperature applications such as gas turbines, combustion cans, ducting, and structural components for power-generating plants [7]. Numerous studies on Alloy 617 have been performed to investigate creep properties, tensile behavior at various

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temperatures, corrosion resistance, and thermal aging effects [8–22]. In 1974, Mankins et al. studied the aging effects on the microstructure and phase stability of Alloy 617 at 649–1093 °C and identified $M_{23}C_6$ as the main stable precipitates at all temperatures and a small amount of gamma prime ($L1_2$-structured Ni_3Al) at 760 °C [9]. The aging study of Alloy 617 was extended to 8000 h in the temperature range of 593–816 °C by Kimball et al., and $M_{23}C_6$ carbides precipitating at grain boundaries were again reported [11]. Later in 1980, Kihara et al. studied the creep properties at 1000 °C and discussed the microstructural development, especially the evolution of $M_{23}C_6$ precipitates during creep, and proposed the existence of the Ostwald ripening mechanism under loading [12]. Recently, Mo et al. studied the aging effects at 900 °C and 1000 °C up to 3000 h, using electron microscopy techniques to provide detailed information on the size distribution, morphology, and coherency of $M_{23}C_6$ precipitates [13–15]. Ren et al. have reviewed the aging effects on Alloy 617, and summarized most of the previous works and important experimental data [16,17].

In previous studies, $M_{23}C_6$ has been identified as the major precipitate during high-temperature thermal aging, and has been considered the main reason for changes in strength/hardness of Alloy 617 [13]. The size, volume fraction, and location of the precipitates are dependent on the thermal aging conditions. To better understand their behavior and interaction mechanism with the metallic matrix during mechanical loading in different aging conditions, we conducted *in-situ* tensile tests combined with synchrotron wide-angle X-ray scattering (WAXS) to characterize the microstructural changes during loading. The experiments are based on long-term (up to 10,000 h) thermal aging experiments on Alloy 617 at 1000 °C [14,15,22]. From the high-energy X-ray measurements, both the macroscopic tensile behavior and the microstructural evolution involving the change in volume fraction of the carbide phase, the lattice strain development for both carbides and metallic phases, and dislocation densities during loading were determined.

2. Description of experiments

The Alloy 617 was provided by Haynes International, Inc. in plate form. The one-inch thick plate has a nominal composition of Ni–22.1Cr–12.2Co–9.46Mo–1.104Fe–0.39Ti–1.03Al–0.064Mn–0.08C–0.05Si–0.017Cu, in wt%. The alloy was hot worked and solution treated at 1177 °C for 37 min. The heat number was 861758808 [13]. In order to study the thermal aging of Alloy 617 for applications in the IHX component of VHTR, the aging experiments were performed in laboratory air conditions at 1000 °C, which was above the gamma prime solvus [9]. The alloy was aged to 10, 30, 100, 300, 1000, 3000, and 10,000 h so that the aging effects could be systematically characterized. Miniature tensile specimens were then machined from the aged alloys. The effects of aging on tensile behavior were investigated by performing *in-situ* tensile tests and WAXS at room temperature. The experiments were carried out at the 10-ID-B beamline at the Advanced Photon Source (APS), Argonne National Laboratory (ANL).

The synchrotron WAXS experimental setup is shown in Fig. 1. The tensile tests were performed with a Deben 5 kN dual lead-screw tensile stage. The *mar*345 image plate X-ray detector was used to measure X-ray diffraction patterns. The energy of the incident X-ray beam was 50 keV (λ=0.2480 Å). The dimensions of the miniature tensile specimens were 1.19 mm × 0.75 mm × 5.00 mm (width × thickness × gauge length). Uniaxial tensile tests were performed in the displacement control mode, with a displacement step of 0.02 mm in the elastic region and 0.05–0.10 mm in the plastic region. After each displacement, the position was kept constant during each X-ray exposure. The speed range of the gearbox was in the range of 20 μm/min to 1.5 mm/min, and the highest strain rate was estimated to be around 0.005/s, which was slightly higher than conventional tensile tests that use a strain rate of 10^{-4}/s to 10^{-3}/s. Nonetheless, the tensile data can be compared with existing data that employed different strain rates, since Alloy 617 has been known to be insensitive to strain rate at ambient temperature [23,24]. In order to cover more grains during each exposure to reduce statistical errors, the X-ray beam size was chosen to be 700 μm × 700 μm, and the total diffraction volume of around 0.3675 mm^3 was attained. The X-ray exposure time ranged from 2 s in the elastic region to about 30 s near fracture so as to maximize the signal-to-noise ratio without saturation. Load and displacement data were recorded to determine the macroscopic stress–strain curves. Due to limited beam time, only one sample was tested for each aging time point. *Ex-situ* tensile measurements were carried out independently to determine the uncertainty in the applied stress, which is estimated to be less than 2%.

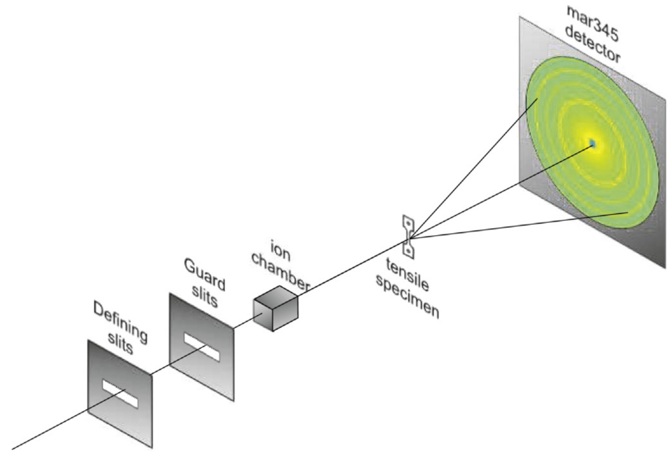

Fig. 1. Schematics of *in-situ* synchrotron X-ray diffraction experiment setup. The energy of incident X-ray beam was 50 keV. A *mar*345 image plate detector was used for X-ray detection.

In order to validate the WAXS results, transmission electron microscopy (TEM) was used to characterize the $M_{23}C_6$ carbides and measure the dislocation densities in the alloy that was aged for 10,000 h. Both pre-tensile and post-tensile miniature samples were examined. TEM specimens were lifted out from the gauge part of the mechanically polished miniature samples using an FEI HELIOS 600i FIB. Bright field (BF) images were used to measure the average dislocation density. Selected area diffraction (SAD) was used to characterize the $M_{23}C_6$ carbides. All the TEM images were taken using a JEOL 2010 LaB_6 TEM operated at 200 kV.

3. Results and discussion

3.1. Macroscopic tensile properties during aging

Macroscopic tensile data are shown in Fig. 2. Fig. 2(a) shows the engineering stress–strain curves for both the as-received and aged specimens. The yield strength (YS) and the ultimate tensile strength (UTS) are shown in Fig. 2(b). Compared to the alloy in the as-received condition, the YS and UTS of the alloy aged for t=10 h increased by 35 MPa and 22 MPa, respectively. However, for the samples aged for 10 h up to 3000 h, only slightly variations in both the YS and UTS have been observed. In the 10–3000 h regime, the average YS and UTS are 450 MPa and 790 MPa, respectively. For the samples aged for 10,000 h, the YS and UTS decreased to 380 MPa and 720 MPa, respectively. This shows that the long-term