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ScienceDirect

Acta Materialia 83 (2015) 507–515

Acta MATERIALIA

www.elsevier.com/locate/actamat

Effect of short-range ordering on stress corrosion cracking susceptibility of Alloy 600 studied by electron and neutron diffraction

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Received 26 July 2014; revised 6 October 2014; accepted 7 October 2014
Available online 15 November 2014

Abstract—Slow strain rate tests (SSRTs) were conducted on low-temperature mill-annealed Alloy 600 tubes at 250 and 360 °C in water with either 8 ppm or below 10 ppb of dissolved oxygen (DO). A special tensile specimen design with a hump was employed for these tests. During SSRT in 360 °C water, stress corrosion cracking (SCC) or intergranular cracking of Alloy 600 is enhanced at DO contents below 10 ppb but suppressed at 8 ppm DO. The SCC susceptibility of Alloy 600 is observed to be related to the degree of lattice contraction by short-range ordering, which is enhanced in the presence of hydrogen. By analyzing electron and neutron diffraction patterns before and after SSRT in 360 °C water, for the first time, definitive evidence is presented for the short-range ordered phase with a d-spacing of 2.1 Å being formed in Alloy 600 during SSRT in 360 °C water, which is manifested by the forbidden reflections at the 1/3{422} positions in ⟨111⟩ selected area diffraction patterns.

Keywords: Stress corrosion cracking (SCC); Intergranular (IG) cracking; Lattice contraction; Short-range ordering (SRO); Alloy 600

1. Introduction

Stress corrosion cracking (SCC) of structural materials made of Alloy 600, such as steam generator tubes, heater sleeves and various nozzles, is a hot issue in the nuclear industry. Given the main features of SCC where cracks grow primarily by intergranular (IG) cracking [1,2], several hypotheses have been proposed for several decades assuming that IG cracking is an extrinsic phenomenon that is governed by degradation of grain boundaries by either corrosion [3,4] or oxidation [5,6]. In contrast, IG cracking of austenitic Ni–Cr–Fe alloys was observed to occur even under an inert atmosphere such as argon, where no corrosion or oxidation of the grain boundary is involved [7,8]. These observations suggest that IG cracking may be an intrinsic phenomenon which is against the current hypotheses related to SCC of metals. Another misunderstanding of SCC is the residual tensile stress, which has been regarded as the primary factor leading to earlier SCC failures of the welds and nozzles of Alloy 600. However, if this residual stress is the cause of SCC, the IG cracking of the welds should occur more frequently at the beginning of life, and not after a long incubation of ~100,000 h [9]; the reason for this remains unresolved so far. In fact, considering that the residual stress is relieved by 80–100% within 24 h at a temperature of 300 °C and above [10], the residual tensile stress cannot account for the occurrence of SCC after such long incubation times. Nonetheless, this observation that a long incubation time is required prior to SCC initiation in the structural components made of Alloy 600 suggests that stresses are internally built up in Alloy 600 by an internal factor during a sufficiently long incubation time that are high enough to exceed the critical stress for IG cracking. In other words, IG cracking of Alloy 600 is dictated by an internal mechanism, which is our unique idea. As a matter of fact, Marucco [11] observed that the lattice contractions of up to 0.12% occurred in several nickel alloys due to short-range ordering (SRO) upon aging at 475 °C for 32,000 h. Nevertheless, she did not appreciate the effect of the lattice contractions by SRO on SCC or IG cracking. When the grains are contracted due to SRO, however, the grain boundary would be subjected to tensile stresses that lead to mechanistic cracking of grain boundaries or IG cracking or build-up of higher strains or dislocations in the regions adjacent to the grain boundaries in the case where IG cracking is absent. Support of this suggestion is provided by Kohara and Kuzynski's observation [12] that a tricrystal of CuAu broke into three pieces by IG cracking 35 seconds after an ordering reaction at 350 °C, and Hou et al.'s observations [13,14] of higher strains or a higher number density of dislocations in the regions adjacent to the grain boundaries. Thus, the more enhanced IG cracking of austenitic Ni–Cr–Fe alloys in water than in argon [7] and the hydrogen-enhanced IG cracking in Alloy 600 [15–17] appear to be related to the enhanced ordering by hydrogen, as already revealed by Flanagan et al. [18].

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http://dx.doi.org/10.1016/j.actamat.2014.10.009
1359-6462/

Considering that atoms in materials keep changing their positions by either plastic deformation or thermal diffusion, SRO leading to lattice contractions cannot be avoided in alloys with several solute atoms during their use, especially at high temperatures, which is the cause of SCC of structural materials, according to our proposal. The aim of this work is to demonstrate that SCC of Alloy 600 is related to SRO. To this end, slow strain rate tests (SSRTs) were conducted in recirculating water at either 250 or 360 °C on an Alloy 600 tube which had been subjected to low temperature mill annealing. To show if the environmental effect on IG cracking of an Alloy 600 tube is related to a hydrogen-enhanced SRO, the content of dissolved oxygen in water was varied from below 10 ppb to 8 ppm (i.e. by about three orders of magnitude). The lattice spacing was determined before and after SSRT using neutron diffraction to correlate IG cracking susceptibility of Alloy 600 with SRO. Direct evidence for the short-range ordered phase being formed in Alloy 600 during SSRT in 360 °C water is presented by comparing electron and neutron diffraction patterns before and after SSRT.

2. Experimental procedures

An Alloy 600 tube mill annealed at 960 °C for 10 min followed by water quenching was used in this study. This low-temperature mill-annealed (LTMA) tube has a typical chemical composition of Alloy 600 with a low carbon content of 0.01 wt.%, as shown in Table 1, which is in accordance with ASTM B-167. SSRT was conducted on tensile specimens with a hump using a constant extension rate test (CERT) machine and a water loop at a strain rate of $2.5 \times 10^{-7}\ \mathrm{s}^{-1}$. The hump specimens, as shown in Fig. 1, were used to enhance IG cracking in the LTMA Alloy 600, which is herein called Alloy 600. During SSRT, the specimens were exposed to either 250 or 360 °C in pressurized water to 200 atmospheres and containing either 8 ppm or less than 10 ppb of dissolved oxygen (DO). More detailed procedures are given elsewhere [19]. After SSRT, the fracture surfaces of the specimens were examined by scanning electron microscopy (SEM) to observe the cracking pattern and determine the fraction of IG cracking. Transmission electron microscopy (TEM) thin foils were obtained by a focused ion beam from a fractured surface of the tensile specimen and were examined in a JEM 2010 microscope (JEOL, Japan) at 200 kV to obtain images and electron diffraction patterns at the leading edge of a crack that was grown either along the grain boundary or into the grains. The diffraction patterns were obtained from the [1 1 1], [1 1 0] and [1 1 2] zone axes to confirm if diffuse peaks generated by short-range order is a fact or an artifact. A neutron diffraction analysis was conducted using a neutron beam wavelength of 1.837225 ± 0.000034 Å on the gauge sections of the tensile specimens before and after SSRT. The instrumental resolution of the diffractometer was $\Delta d/d \cong 0.04\%$. The change in peak position (peak shift) was used to determine a change in lattice parameter and its sensitivity was less than 1/10th of the instrumental resolution.

Table 1. Chemical composition of an LTMA Alloy 600 tube used.

Alloy 600	Ni	Cr	Fe	Mn	C	Cu	Si	S
wt.%	75.1	15.4	8.0	0.3	0.01	0.2	1.0	0.001

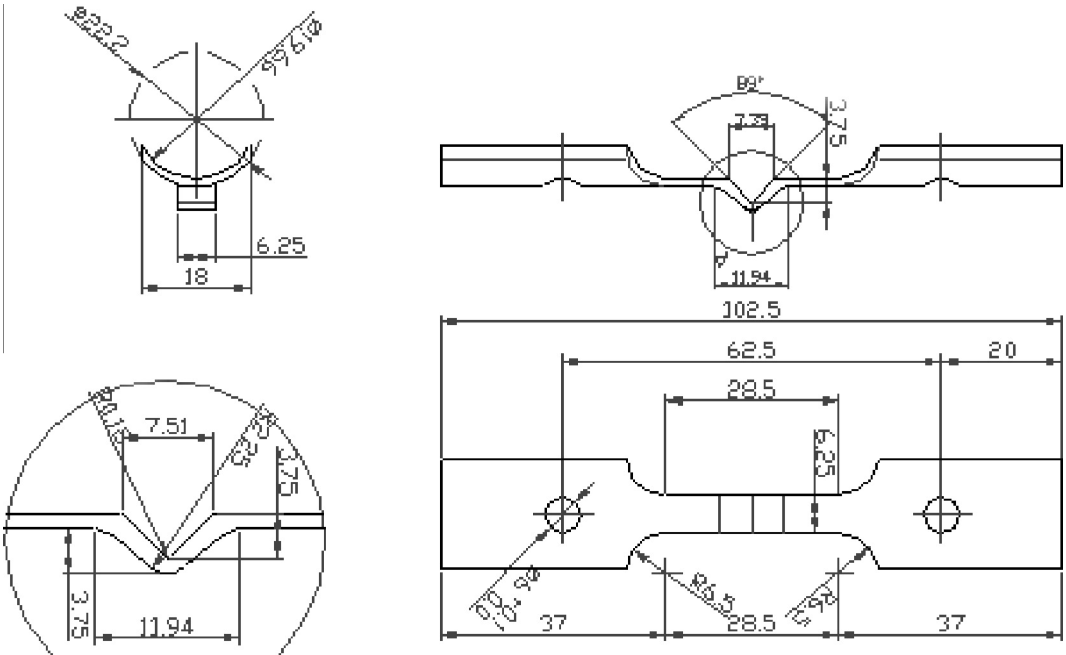

Fig. 1. Dimension of tensile specimens with a hump used for slow strain rate tests.