Journal of Nuclear Materials 466 (2015) 443–459

Contents lists available at ScienceDirect

Journal of Nuclear Materials

journal homepage: www.elsevier.com/locate/jnucmat

Analysis of stress corrosion cracking in alloy 718 following commercial reactor exposure☆

Keith J. Leonard [a,*], Maxim N. Gussev [a], Jacqueline N. Stevens [b], Jeremy T. Busby [a]

[a] Oak Ridge National Laboratory, Oak Ridge, TN, USA
[b] AREVA Inc., Lynchburg, VA, USA

ARTICLE INFO

Article history:
Received 20 February 2015
Received in revised form
6 August 2015
Accepted 22 August 2015
Available online 24 August 2015

Keywords:
Alloy 718
Post-irradiation examination
Stress corrosion cracking

ABSTRACT

Alloy 718 is generally considered a highly corrosion-resistant material but can still be susceptible to stress corrosion cracking (SCC). The combination of factors leading to SCC susceptibility in the alloy is not always clear enough. In the present work, alloy 718 leaf spring (LS) materials that suffered stress corrosion damage during two 24-month cycles in pressurized water reactor service, operated to >45 MWd/mtU burn-up, was investigated. Compared to archival samples fabricated through the same processing conditions, little microstructural and property changes occurred in the material with in-service irradiation, contrary to high dose rate laboratory-based experiments reported in literature. Though the lack of delta phase formation along grain boundaries would suggest a more SCC resistant microstructure, grain boundary cracking in the material was extensive. Crack propagation routes were explored through focused ion beam milling of specimens near the crack tip for transmission electron microscopy as well as in polished plan view and cross-sectional samples for electron backscatter diffraction analysis. It has been shown in this study that cracks propagated mainly along random high-angle grain boundaries, with the material around cracks displaying a high local density of dislocations. The slip lines were produced through the local deformation of the leaf spring material above their yield strength. The cause for local SCC appears to be related to oxidation of both slip lines and grain boundaries, which under the high in-service stresses resulted in crack development in the material.

1. Introduction

The performance of Ni-base alloy 718 in light water reactor systems is dependent on many factors that include the starting microstructure and its subsequent evolution under the combination of applied stresses, irradiation and corrosive environments. While generally considered resistant to primary-water stress corrosion crack (SCC) initiation, alloy 718 can be susceptible to crack propagation. The starting microstructure of the material and the changes it undergoes are directly linked to the mechanical strength and corrosion properties of the component and influence the susceptibility of the material to crack propagation mechanisms leading to degraded function or failure of components. While Alloy 718 is used in numerous significant nuclear and nonnuclear applications, the crack propagation behavior and conditions that promote SCC in alloy 718 are not as well studied as those of other alloys, such as austenitic steels. This is specifically the case for material that has shown failure during reactor exposure.

Damaged cruciform-type hold-down leaf spring (LS) arms were observed during refueling outage inspections at several pressurized water reactors (PWRs) between 2007 and 2009. Two sets of LS assemblies from a US PWR reactor were harvested from the upper end fitting of the fuel assembly after two 24-month cycles in the reactor operated to >45 MWd/mtU burn-up. The assemblies were shipped to the Irradiated Materials Examination and Testing hot cell facility at Oak Ridge National Laboratory (ORNL) for inspection and disassembly and to be cut into test materials. Further in-depth studies on smaller components cut from select sections of the leaf

☆ *This manuscript has been authored by UT-Battelle, LLC, under Contract No. DE-AC05-00OR22725 with the U.S. Department of Energy.* The United States Government retains and the publisher, by accepting the article for publication, acknowledges that the United States Government retains a non-exclusive, paid-up, irrevocable, world-wide license to publish or reproduce the published form of this manuscript, or allow others to do so, for United States Government purposes. The Department of Energy will provide public access to these results of federally sponsored research in accordance with the DOE Public Access Plan (http://energy.gov/downloads/doe-public-access-plan).

* Corresponding author. Materials Science and Technology Division, Oak Ridge National Laboratory, P.O. Box 2008, MS-6138, Oak Ridge, TN 37831-6138, USA.
E-mail address: leonardk@ornl.gov (K.J. Leonard).

http://dx.doi.org/10.1016/j.jnucmat.2015.08.039
0022-3115/

spring assemblies were performed at the ORNL Low Activation Materials Development and Analysis (LAMDA) laboratory.

In the post-irradiation examination work reported herein, we examine the crack propagation characteristics and statistical analysis as related to the route of propagation. Evaluation of potential crack nucleation sites and characterization of the cracks were also performed through transmission electron microscopy (TEM) and is discussed with general microstructural and hardness property differences between the irradiated and nonirradiated (archived) samples available from the same fabrication processing batch of material.

2. Material and experimental methods

The alloy 718 spring material is cold rolled, machined, solution annealed and precipitate hardened through heat treatments in accordance with the Aerospace Materials Specification 5596 [1] and LS vendor's internal proprietary specifications. The alloy composition is listed in Table 1.

For the ~45 MWd/mtU burn-up that the LS assemblies received during the two 24-month cycles in PWR service, the irradiated material presented in this study received a dose of 9×10^{23} n/m^2 ($E > 1$ MeV). Equivalent displacement damage based on experimental reactor conditions assuming a primary influence of fast neutrons would produce approximately 0.14 displacements per atom (dpa) [2]. Taking into account thermal and epi-thermal neutrons, the effective damage level could be a little higher due to displacement damage generated through $Ni^{59}(n,\alpha)Fe^{59}$ reactions [3]. He generation is not considered to be substantial due to the low total thermal fluence and B content within the alloy. Irradiation temperature during reactor service was nominally 317 °C.

Analysis of cracked features and polished metallographic samples was conducted using a Keyence VHX-1000 digital confocal microscope with image stitching capability, allowing for the large-area investigations. Vickers microhardness data (500 g load, 10 s dwell time) was collected on either etched or electropolished surfaces of prepared metallographic samples. Tests performed on archival materials in the polished and etched conditions revealed no statistical difference.

Electron-backscatter diffraction analysis (EBSD) was performed using a field emission JEOL JSM 6500F scanning electron microscope (SEM) equipped with an orientation imaging microscopy (OIM) system by EDAX. For EBSD work, the SEM accelerating voltage was 20 kV; the working distance varied from 12 to 15 mm. The EBSD maps were measured on a hexagonal grid with a step size of 0.1–2 μm. Each of the beam-scan fields was measured at a magnification of 5000× or more to ensure that all points were in focus.

All EBSD data were filtered (minimum grain size three points, maximum point-to-point misorientation 5°) to reduce indexation errors. Image quality (IQ) maps were drawn to obtain qualitative information about the microstructure of the deformed samples. The IQ maps describe the quality of the electron backscatter patterns, which is related to the strain distribution in the microstructure [4] as well as reveal details such as grains, twins, phase boundaries, cracks, and slip bands.

Samples for TEM were prepared either through conventional twin-jet electropolishing of 3 mm discs from the archive material, or through focused-ion-beam (FIB) milling of the irradiated material. Jet-polished samples were prepared using a Struers Tenupol polisher with an electrolyte of 10 vol % $HClO_4$ in methanol, at temperatures between −20 to −30 °C and ~20 V. Specimens for TEM from the irradiated material were prepared by FIB milling using an FEI Quanta 3D 200i instrument at site-selective locations, which included lift-out samples from the cracked regions and representative lift-outs from the bulk material away from the cracks. Samples were examined on an FEI CM200 TEM, a Schottky 200 keV field emission instrument with scanning-TEM (STEM) capabilities and equipped with an energy-dispersive spectrometer (EDS), a high-angle annular dark-field (HAADF) detector, and a Gatan image filter (GIF) for electron energy loss spectroscopy (EELS).

For the as-received specimens, the stress corrosion cracks at the surface were, in most cases, partially filled by corrosion products, restricting the view of the cracked surfaces and their analysis. A cleaning and etching procedure was employed to remove oxide layers and sediments and to get a clear metallic surface. The procedure used was as follows:

1. A 5 min soak at ~100 °C (boiling) solution of 30 g/L potassium permanganate and 100 g/L sodium hydroxide.
2. An additional 5 min soak in ~100 °C (boiling) solution of 30 g/L ammonium oxalate.
3. Ultrasonic cleaning of the specimen in distilled water for up to 3 min.
4. Repeating steps 1–3 if found necessary.

The cleaning procedure was repeated up to six times to provide appropriate results; after cleaning, the oxide layer at the surface and the oxide within the near-surface cracks were fully removed. For comparison and control reasons, the same cleaning procedure was performed on an archive specimen of the same alloy (no corrosion layer). It was found that the control specimen did not reveal any change or damage to the surface in the form of etching, pitting, or overall material loss from the cleaning procedure, providing confidence that the structures revealed in the irradiated material following this procedure are an accurate representation of the in-service material.

3. Results and discussion

3.1. General appearance of SCC in the material investigated

Fig. 1 provides examples of typical damage observed in the LS material. The optical images in this figure are composites of images taken across the width of the leaf arm at the location of maximum stress near the base of the arm located near the retaining nut (no cracking was observed at lower stress regions). Details regarding LS design and fabrication are proprietary. The level of in-service tensile stress at the cracked area, estimated using finite element analysis (FEA), was found to be ~1000–1200 MPa. It is difficult to provide more precise estimations because the stress level likely changed both during operation (fuel assembly growth) as cracks initiated and propagated.

For the two LS assembly examples, approximately 20% of the leaf arms showed complete fractures (though some may have occurred during handling and shipping), 50% showed cracking to a varying degree (with examples shown in Fig. 1), and 30% showed no

Table 1
Elemental composition of the material investigated (in wt %).

Ni	Cr	Fe	Nb	Mo	Ti	Al	Si	Mn	C	Co, Ta, Cu	P, S, B
Bal.	17.9	18.0	4.93	2.95	1.05	0.44	0.07	0.08	0.028	<0.008	<0.002