Journal of Nuclear Materials 296 (2001) 139–144

www.elsevier.com/locate/jnucmat

The mechanical properties of an Alloy 718 window after irradiation in a spallation environment

M.R. James [a,*], S.A. Maloy [a], F.D. Gac [a], W.F. Sommer [a], J. Chen [b], H. Ullmaier [b]

[a] *Los Alamos National Laboratory, P.O. Box 1663, MS H809 Los Alamos, NM 87545 USA*
[b] *Forschungszentrum Jülich, Projekt ESS, D-52425 Jülich, Germany*

Abstract

A double-shelled Alloy 718 window was irradiated at the Los Alamos Neutron Science Center (LANSCE) with a 1 mA, 800 MeV proton beam to a maximum dose of 20 dpa at a calculated irradiation temperature of 400°C. After irradiation, the window was visually inspected in hot cells at the Chemical and Metallurgical Research (CMR) facility at the Los Alamos National Laboratory (LANL) and cut into strips for analysis. Microhardness and optical metallography were performed at the LANL hot cells and tensile testing and gamma measurements were performed at Forschungszentrum Jülich. The results of these analyses are presented in comparison with previous results obtained on similarly irradiated Alloy 718 components. A crack that occurred in-service was also examined and possible failure modes are discussed. © 2001 Elsevier Science B.V. All rights reserved.

1. Introduction

During operation of the Los Alamos Neutron Science Center (LANSCE) accelerator an Alloy 718 window was exposed to a beam of high-energy protons and reached a peak dose of approximately 20 dpa. The window was removed from service after the 1996–1997 irradiation campaign and visually inspected following coolant leaks that occurred in-service. Strips of material were also cut out of the window and sent to Forschungszentrum Jülich (FZJ) for mechanical testing. At FZJ, tensile samples were cut from these strips and tested at room temperature. Additionally, hardness tests and metallographic examination were performed at Los Alamos National Laboratory (LANL). It has been shown previously that precipitation-hardened (PH) Alloy 718 exposed to high-energy (800 MeV) proton and spallation-neutron radiation at temperatures from 50°C to 160°C undergoes changes in mechanical properties that are similar to the changes seen in materials exposed to lower energy neutron (i.e., fission) environments at the same temperature up to $\sim$1 dpa [1]. The window material was exposed at higher temperatures (367–400°C) and a much higher dose than these previous studies. The results of these tests are presented in this work and comparisons will be drawn to similar testing.

2. Window design

The LANSCE beam window is a hemispherical double-shell design with cooling water flowing between the two shells, see Fig. 1. The outer hemispherical shell is 2.36 mm thick and has an outer radius of 95.25 mm. The inner shell is 2.24 mm thick with an outer radius of 91.19 mm. A nominal separation of 1.70 mm between the shells is maintained by a series of wires [1] tack-welded to the outer surface of the inner hemisphere. The wires also form a serpentine path for the coolant but

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[*] Corresponding author. Tel.: +1-505 665 6521; fax: +1-505 667 2787.
E-mail address: mrjames@lanl.gov (M.R. James).

[1] The wire material used in the window fabrication was not recorded although the specifications call for 316 SS or Alloy 625. Analysis of the wire material is planned.

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PII: S0022-3115(01)00516-5

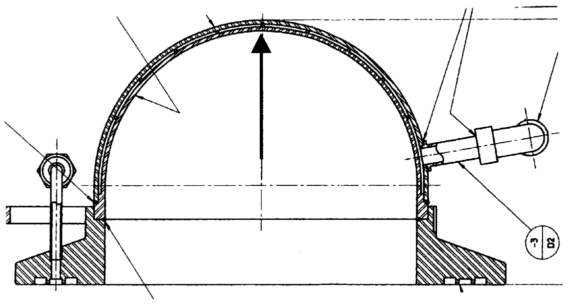

Fig. 1. LANSCE beam window cross-section. The arrow describes the traverse of the proton beam.

because their diameter is only 1.59 mm, some cross-flow occurs and turbulence from this flow decreases the coolant efficiency. The two shells were machined from solid blocks of Alloy 718 and heat treated to the PH condition. The window serves as a barrier between the vacuum of the accelerator components and the target area, which is kept at atmospheric pressure. The window is heated from the passage of the proton beam and is also pressurized by the cooling water. Thermal–hydraulic analysis of the operation of the beam window [2] estimates the maximum von Mises stress at 606 MPa and the nominal peak operating temperature of the window material between 367°C and 400°C [3]. However, there are strong indications that the coolant flow was in a two-phase regime that could have led to even higher temperatures. While there were thermocouples affixed to the exterior surface of the window, the readings obtained were hundreds of degrees higher than the temperature of the window due to proton beam heating effects.

3. Irradiation history

The beam window under discussion was installed in the LANSCE accelerator in April 1995. From that time until it was removed from service in 1997, it was exposed to approximately nine months of full power (1 mA) operation (the longest service of any LANSCE beam window). A leak in the window was first noticed after about six months of operation from an increase in radioactive isotopes in the atmosphere of the target region. The source of the leak was confirmed from pressure tests on the window coolant circuit. The vacuum (accelerator) side was not compromised so the window was allowed to remain in service until the end of the irradiation campaign. Also, during the last months of operation and coincident with the leak in the window, the LANSCE accelerator began to suffer numerous, short-term trips from sparking in the proton source. This resulted in many thermal cycles on the window. The number of cycles was 2 per h in the first two months of operations and in the last four months of operation was approximately 4 per h amounting to approximately 15 000 cycles.

4. Post-irradiation examination

After the window was removed from the accelerator, a plate containing radio-chromic film was exposed on the top surface. Tungsten wires placed between the film and the window marked the geometric window center. The film's sensitivity to radiation revealed that the highest activity region corresponded to the window center, confirming the beam alignment. The window was shipped to the hot cell facilities at the Chemical Metallurgical Research (CMR) Facility at Los Alamos for further examination.

Disassembly of the window immediately revealed a scale adhering to the surfaces between the two shells in the vicinity of the beam center (see Fig. 2). The deposit was later determined to be copper in the form of a copper oxide (tenorite, CuO). The presence of copper in the cooling water was not unexpected as the same water loop is used to cool the copper beam stop. The deposition, in conjunction with other evidence including thermal–hydraulic analysis of the window [3], strongly suggests that boiling was occurring at this point in the window. Another feature of interest was the degradation of the spacer wire that separated the two shells. The wire that ran across the window center (and therefore the beam center) showed a reduction of cross-sectional area and a fracture near the window center. Disruption of the coolant flow by the serpentine arrangement of the wires, tack welds that held the wires and possible cross-flow conditions all served to exacerbate the hot spot and lead to boiling of the water and poor cooling in the window center.

Examination of the window after disassembly revealed a crack in the outside shell located ~1 cm from the center, a location coincident with the copper deposit and close to the center spacer wire. The crack length was about 0.5–1 cm and branched in at least two locations. The crack, shown in Fig. 3, was also observed to be close to spot welds (resistance welds) that had been used to attach thermocouple pads to the outside of the window surface. These spot welds were applied following the heat-treatment of the window to the PH condition. While in some cases of high heat-load welding, liquidus cracking can be induced in PH Alloy 718, a greater concern is strain-age cracking [4]. Strain-age cracking can occur when the alloy is welded (in either the annealed or PH condition) and the residual stresses in the weldment are not relieved by annealing prior to placing the component in elevated temperature service. In this unannealed condition, aging in the presence of residual

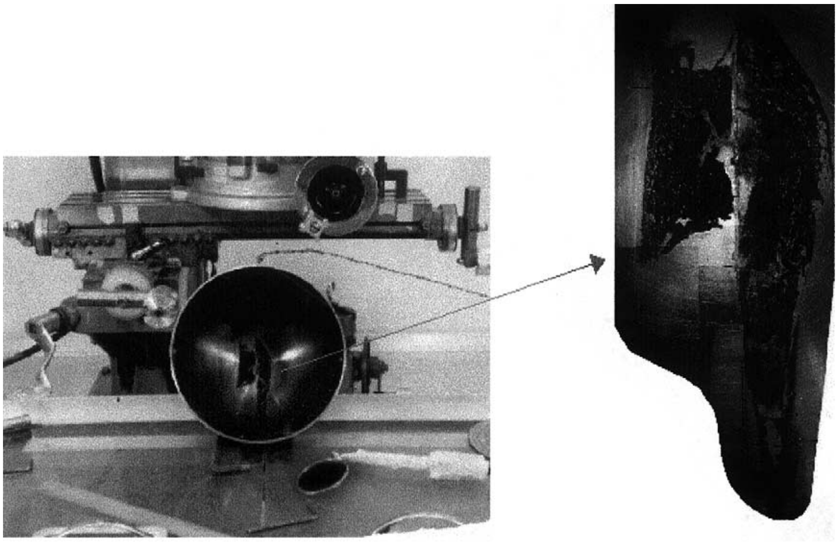

Fig. 2. Images of deposit seen on interior cooling channel.

stresses can result in cracking of the material in the weld heat-affected zone. It is for this reason that it is recommended that Alloy 718 be re-annealed and re-aged following welding for service in temperatures in the age-hardening regime [5]. The threshold aging temperature, where strain-age cracking might occur, is usually between 540°C and 650°C, although there are suggestions that a weld can age-harden at temperatures as low as 425°C [6]. Given the degraded flow conditions, it is very likely that the peak window temperatures during service could have been sufficient for strain-age cracking to occur. The effects of the proton-beam irradiation could also have served to accelerate the aging [7] or reduce the yield strength and/or ductility of the material in this region.

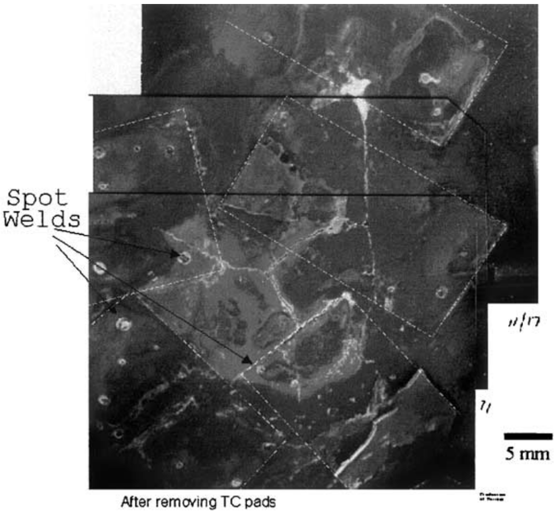

Fig. 3. Image of crack as seen from the outer surface. The locations of the thermocouple pads are given by the white-dashed lines. The spot welds were the attachment points for thermocouple pads.

Other environmental factors could also have contributed to the crack including (1) the harsh corrosion environment on the outer surface from the formation of nitric acid in the target area and (2) energy deposition in the thermocouple pads may also have contributed to local hot spots.

As noted earlier, the accelerator continued operation after the initial cracking. The rate of cooling water leakage was monitored and did not increase substantially during the final three months of operation. This implies the crack did not grow appreciably despite the numerous thermal cycles and loss of elongation (and possible embrittlement) of the Alloy 718. Further examination of the crack surface is planned to gather more information on its origin and growth.

5. Future window designs

The observations made on the irradiated LANSCE window led to important changes in subsequent window designs: (1) Thermocouples were not attached to the window since the spot welds were implicated in the cracking in the previous window. (2) The location of the spacer wires and flow channels was also changed to prevent a hot spot resulting from the center of the beam being coincident with a spacer wire. Improvements were also made in the performance of the proton source at LANSCE that served to greatly minimize the number of beam trips in subsequent irradiations.

6. Material analysis

Strips were cut out of the inner shell of the window using a slitting saw on a milling machine and were used in analysis of the post-irradiation materials properties. The dimensions of the strips were 15 mm × 4 mm × 2.23 mm. These were sent to FZJ where gamma analysis was performed for retrospective dosimetry. The results of these analyses were compared to gamma analysis on Alloy 718 samples which had been irradiated in the LANSCE proton beam and for which doses had already been determined from activation foils [8]. Evaluation of the isotopes, primarily Mn-54, revealed the highest dose sample from the window reached approximately 20 dpa. The strips were mounted and sliced such that the 4 mm dimension was divided into two samples of roughly 0.6 mm thickness each. The final strips were rectangular samples approximately 0.6 mm thick, 2.2 mm wide and 15 mm long. Dog-bone tensile samples were then fabricated from these strips using a polishing wheel. The tensile samples were then tested at an initial strain rate of 1×10^{-3} s^{-1} at room temperature on an MTS test machine. Elongation data were taken from a video camera that monitored the gauge section of the tensile sample. Slices from the window were also mounted, polished to a 1 μ diamond finish and etched for metallographic analysis.

7. Results and discussion

7.1. Metallography

A representative metallograph from the center of the window is given in Fig. 4. Comparisons to other locations that were prepared under the same polishing conditions, suggest a possible darkening of the grain boundaries as one approaches the center of the window, where the proton beam was most intense. The darkening is hypothesized to be from a second phase forming at the grain boundaries as has been observed in previous studies of neutron-irradiated Alloy 718 at 288°C [9].

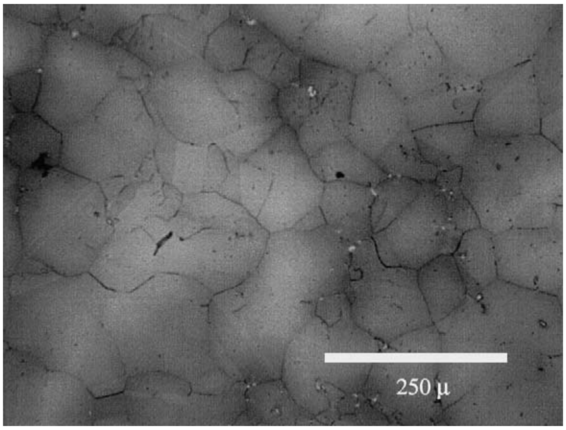

Fig. 4. Polished Alloy 718 from the center of the window.

7.2. Tensile tests

The results of the tensile tests on the Alloy 718 taken from the window are shown in stress–strain curves given in Fig. 5. In this figure, the tendency for the Alloy 718 to increase slightly in strength can be readily seen. Also shown is the drastic reduction in the elongation as a function of dose. In particular, the 20 dpa test failed with zero elongation and the result is visible in Fig. 5 as the precipitous load drop just after the elastic portion of the curve.

The changes in yield strength (YS) as a function of dose are shown in Fig. 6. In the same figure are the

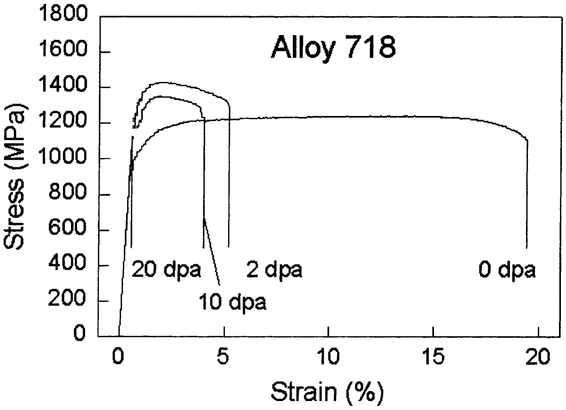

Fig. 5. Stress–strain curves from tensile tests on Alloy 718 extracted from the LANSCE beam window. All tests were performed at room temperature.

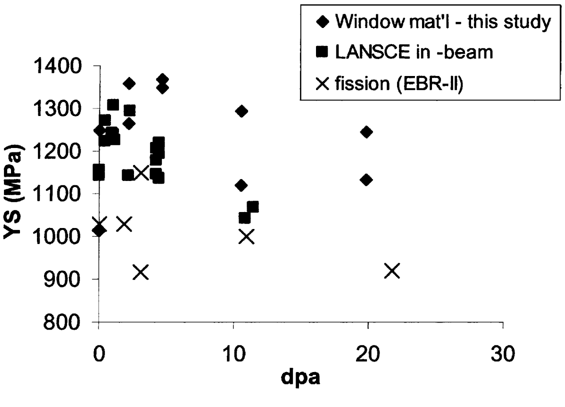

Fig. 6. YS vs. dose for the current tests (◆), previous LANSCE-irradiated materials (■) and neutron-irradiated Alloy 718 from EBR-II (×).

results from tensile tests performed using small-scale Alloy 718 tensile samples irradiated at the LANSCE accelerator at 50–160°C [1], and tensile test results from reactor irradiations on Alloy 718 performed at EBR-II [10]. The EBR-II data were performed with miniature buttonhead tensile samples (28.6 mm gauge length and 3.18 mm diameter) and were reported in terms of neutron fluence. To make a comparison with the current results, the results from Ward et al. [10] were converted to dpa using a typical EBR-II spectra taken from Jacqumin et al. [11]. Analysis of this spectra with the dpa cross-section for Alloy 718 revealed a spectral averaged cross-section of 331 dpa-b. The EBR-II samples were irradiated at 400–700°C and tested at a variety of temperatures. Test results from room temperature (and 232°C for total elongation (TE)) are shown for comparison.

Comparison of the three data sets is complicated by the somewhat different conditions employed in each test and the scatter in the data. But it is possible to discern some general trends. The EBR-II results reported by Ward et al. [10] were irradiated from 2 to 22 dpa with a trend of increasing irradiation temperature (400–700°C) with dose. A subset of the results (the samples tested at room temperature) is shown in Fig. 6. Ward and colleagues observed the maximum strength was reached in the lower dose and lower temperature irradiations and concluded that Alloy 718 underwent changes by competing mechanisms of irradiation hardening and thermal overaging such that the higher dose samples reversed the hardening seen at lower dose samples. The LANSCE data reported in Maloy et al. [1] observed the trend of maximum strength in Alloy 718 also at lower doses (~2–3 dpa) with reductions in the YS occurring past 3 dpa. The LANSCE irradiations were performed in a proton beam, with concomitantly high levels of He and H production and at relatively low temperatures (<100°C). Thermal overaging seems unlikely in the case of the LANSCE irradiations; the competing (softening) mechanism may be a radiation-induced disordering of the precipitates. The current tensile data acquired from the LANSCE beam window have temperature exposures that are in between the LANSCE and EBR-II results and despite the scatter in the data, support the general interpretations previously derived for the behavior of Alloy 718. The maximum observed strengths are seen around 2 dpa with reductions in strength at the higher doses.

A plot that compares the TE data from these data sets is given in Fig. 7. The EBR-II data show very little change in TE (although higher dose samples *tested* at higher temperatures do show reductions in elongation), while the LANSCE-irradiated results show a significant drop in TE below 1 dpa and then a gradual decrease with increasing dose. The results also reveal that at doses >20 dpa there is still ~10% elongation for the irradiation conditions in EBR-II. The material from the LANSCE-irradiated window exhibits zero elongation (the samples failed in the elastic region) at 20 dpa. The elongation differences may be attributed to different temperature conditions, the presence of gas (He/H) in the material or even the difference in the type and size of tensile samples used. The embrittlement of the Alloy 718 from the beam window is not fully understood. With the high levels of He and H generated from the proton beam combined with degraded cooling, it is possible that the window reached the temperature necessary for He embrittlement. The formation of grain boundary phases that led to intergranular fracture is also a possibility. Further study into the failure modes will be conducted.

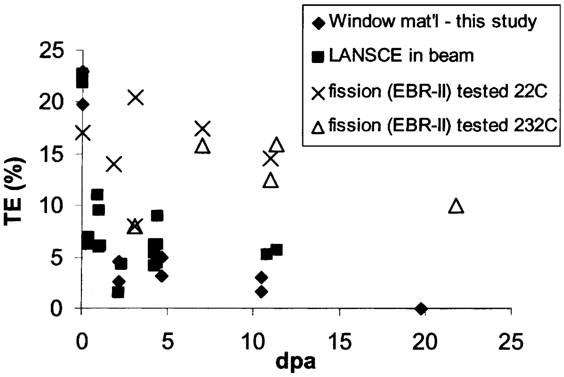

Fig. 7. TE vs. dose for the current tests (◆), previous LANSCE-irradiated materials (■) and fission-irradiated Alloy 718 from EBR-II (×).

7.3. Hardness results

Hardness tests were performed across the strips from ~40 mm to the window center using Diamond Pyramid Microhardness with a 400 g load. The results from a specimen taken from the window edge (very low dose) are consistent with Alloy 718 in the fully PH condition, see Fig. 8. The results near the window center show a significant decrease in hardness and for the data at the center of the window, the hardness appears to rise again. The character of the indents also changed, the indents at ~30 mm from the beam center showed possible indications of strain localization and the indents at the beam center displayed, in some cases, cracks emanating from the indent, indicative of a very brittle material. The results are consistent with the results seen in the tensile tests in the previous section in that the proton-irradiated areas show a softer material compared to the starting (unirradiated) condition and the material at the very center showed signs of embrittlement (failure in the elastic region in tension).

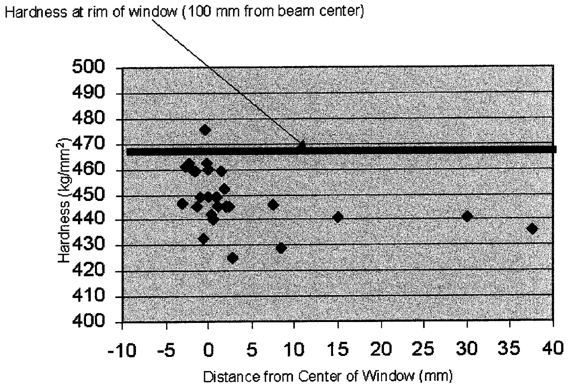

Fig. 8. Hardness measurements across the window. The nominal beam radius was about 20–25 mm. The solid line represents the hardness at the window edge (lowest irradiated condition).

8. Conclusions

A crack occurred in the LANSCE beam window while in service, although it did not compromise accelerator operation. The crack was first detected by the leakage of radionuclides from the window coolant into the target containment box. Post-irradiation examination revealed a crack near the center of the window and in the vicinity of thermocouple pads spot-welded to the outer surface. Possible mechanisms for crack initiation include nitric acid corrosion, additional heating from the in-beam thermocouple pads or strain-age cracking caused by the spot welds or some combination of these. Further examination of the crack surface is planned to better elucidate the initiation and growth mechanisms.

The trends in the tensile data agree with reactor data for changes in YS although the elongation changes differ considerably between the two irradiations with the fission-irradiated samples retaining total elongation to doses >20 dpa compared to the proton-irradiated beam window material failing in the elastic region. The higher irradiation temperature of the EBR-II data (400–700°C compared to 50–400°C for the LANSCE data) and the different tensile samples used make it difficult to isolate the changes in TE strictly to the nature of neutron vs. proton irradiation.

The hardness data were consistent with the tensile data that showed a general softening in Alloy 718 at doses from 5–20 dpa. Cracks observed emanating from the indent suggest a brittle material at the window center.

Acknowledgements

We wish to thank Manuel Lopez and Toby Romero for their contributions in the window disassembly at the hot cells. We also acknowledge the efforts of Jack Kennison in identifying the composition of the copper oxide and Gene Zimmermann for his invaluable assistance in fabricating the tooling and fixtures required for this work. Finally, we acknowledge Bob Johnson for his helpful discussions and comments.

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