Contract No:

This document was prepared in conjunction with work accomplished under Contract No. DE-AC09-08SR22470 with the U.S. Department of Energy (DOE) Office of Environmental Management (EM).

Disclaimer:

This work was prepared under an agreement with and funded by the U.S. Government. Neither the U.S. Government or its employees, nor any of its contractors, subcontractors or their employees, makes any express or implied:

- 1) warranty or assumes any legal liability for the accuracy, completeness, or for the use or results of such use of any information, product, or process disclosed; or
- 2) representation that such use or results of such use would not infringe privately owned rights; or
- 3) endorsement or recommendation of any specifically identified commercial product, process, or service.

Any views and opinions of authors expressed in this work do not necessarily state or reflect those of the United States Government, or its contractors, or subcontractors.

FORGING STRAIN RATE AND DEFORMATION TEMPERATURE EFFECTS ON THE FRACTURE TOUGHNESS PROPERTIES OF TYPE 304L STAINLESS STEEL PRECHARGED WITH TRITIUM

M.J. MORGAN

Savannah River National Laboratory Aiken SC, USA **C. SAN MARCHI** Sandia National Laboratories Livermore CA, USA

N.T. SWITZNER

Colorado School of Mines Golden CO, USA **D.K. BALCH** Sandia National Laboratories Livermore CA, USA

ABSTRACT

Forged austenitic stainless steels are used as the materials of construction for pressure vessels designed to contain tritium at high pressure. These steels are highly resistant to tritium-assisted fracture but their resistance can depend on the details of the forging microstructure. In this study, the effects of forging strain rate and deformation temperature on the fracture toughness properties of Type 304L stainless steel were studied. Forgings were produced from a single heat of steel using four types of production forging equipment - hydraulic press, mechanical press, screw press, and high-energy-rate forging (HERF). Each machine imparted a different nominal strain rate during the deformation. The objective of the study was to characterize the J-Integral fracture toughness properties as a function of the industrial strain rate and temperature. The second objective was to measure the effects of tritium and decay helium on toughness. Tritium and decay helium effects were measured by thermally precharging the as-forged specimens with tritium gas at 340 MPa and 350°C and aging for up to five years at -80°C to build-in decay helium prior to testing. The results of this study show that the fracture toughness properties of the as-forged steels vary with forging strain rate and forging temperature. The effect is largely due to yield strength as the higher-strength forgings had the lower toughness values. Tritium exposures reduced the fracture toughness values remarkably to fracture toughness values averaging 10-20% of as-forged values. Forging strain rate and temperature had little or no effect on fracture toughness properties after tritium exposure.

INTRODUCTION

Tritium reservoirs are constructed from forged stainless steels and filled and stored at the Savannah River Site. The vessels are constructed from forged stainless steels because of their good compatibility with tritium. These steels are highly resistant to, but not immune from, the embrittling effects of hydrogen isotopes and helium from tritium decay. Cracking in storage vessels has been observed after extended service times and material properties like ductility, elongation-to-failure, and fracture toughness are reduced with time as tritium and its radioactive decay product, helium, slowly accumulate within the vessel walls during service [1-4]. Because of tritium aging effects, fracture mechanics properties and steel behavior as a function of tritium and decay helium content are needed for fracture modeling, reservoir life prediction, and safety margin evaluations [5-8].

In this study, the effects, of forging strain rate, forging temperature, and prior annealing on fracture toughness were investigated in forged Type 304L stainless steel. Forging remnants from an earlier forging study were used [9], which included forgings from four different forging processes that spanned two orders of magnitude in the imposed strain rates during forging. Additionally, the role of forging temperature and annealing prior to the final forging step were considered. Previous work considere the effect of hydrogen precharging on the tensile properties of these forging conditions [10]. The study was designed to help answer the question "Which manufacturing process produces the microstructure most resistant to tritium embrittlement effects?"

EXPERIMENTAL PROCEDURE

All forgings used in this study were obtained from prior work and derived from a single heat of steel [9]. The composition of the steel is given in Table 1. Switzner et al. [9] had produced forgings to study the effect of forging strain rate and temperature on microstructure and mechanical properties. Forgings of equivalent dimensions were produced by four different forging processes to achieve a range of forging strain rates: Screw press, mechanical (crank) press, hydraulic press, and high-energy-rate forging (HERF). Two different forging temperatures were considered: 816 or 871°C. Additionally, the effect of annealing at a temperature of 954°C prior to the final forging step was also considered. Thus, forgings with 16 unique processing histories were produced: four forging process, each at two final-forging temperatures, and for each temperature, forged with and without a prior annealing step. The materials and forging processes are summarized in Table 2 and in more detail in Ref. [9].

In brief, all forging was accomplished with material from the same starting bar of type 304L austenitic stainless steel (102 mm diameter, machined to 95 mm diameter prior to forging). The final forging shape was achieved by a threestep process. The two initial extrusion steps (identical for all forgings) reduced the bar to 59 mm diameter. The final upset-forging step resulted in a forged cylinder with diameter of 71 mm. The rate of forging was varied by using different forging equipment for this final upset-forging step; in order of increasing deformation rate: (i) hydraulic; (ii) mechanical; (iii) screw; and (iv) HERF. Nominal deformation rates from Ref. [9] are 1, 5, 10, and 100 strain/s for hydraulic, mechanical, screw, and HERF, respectively.

For this study, arc-shaped fracture mechanics specimens shown in Fig. 1 were machined from the center section of the forgings from Switzner et al. [9]. The specimens were machined from sections of the final forgings previously used for hardness and grain flow characterization, representing the 16 unique processing histories described above.

The specimens were fatigue precracked such that the cracks propagated perpendicular to the cylindrical forging axis (i.e, forging direction) and loaded parallel to the forging axis. Some specimens were pre-charged with tritium at 350°C and 34.5 MPa for 14 days. Decay helium content was developed during storage at -80°C from one to five years prior to testing. The specimen tritium and decay helium concentations were estimated from the measured decay

helium content of a high-energy-rate forged Type 304L specimen given a similar exposure. The tritium exposure conditions are estimated to be sufficient to uniformly saturate the test specimens throughout with a tritium content of approximately 1600 atomic parts per million (appm). Storage at one and five years is estimated to achieve approximately 200 and 600 appm, respectively, of decay helium uniformly distributed in the test specimen. The elastic-plastic Jintegral was evaluated for all specimens at ambient temperature by loading to failure at 0.002 mm/s while monitoring load, load-line displacement and crack extension (using a DC potential-drop technique). Two-to-three tests were conducted for each condition and the data were were analyzed according to ASTM E1820-99. For all test conditions, the requirements for the uncracked ligament and thickness were not satisfied; therefore, all fracture toughness values are reported as unqualified Jo values. While all fracture surfaces showed uniform crack fronts, only tritium-exposed specimens showed no evidence of shear lips along the sides of the specimens (implying plane-strain conditions prevailed for these specimens).

RESULTS

Typical J-R (fracture toughness, J vs. change in crack length, da) curves for the screw press forging process conducted at two temperatures are shown in Fig. 2. The fracture toughness value, J_Q , is given by the intersection of the J-R curve with the 0.2 mm offset line. In general, specimens forged at 816°C had slightly lower toughness values than those forged at 871°C. The J-R curves are steeper for as-forged specimens and flatter for tritium/helium precharged specimens. These figures are typical of all of the four forging processes and forging temperatures.

The average J_Q values for each condition is provided in Table 3. In general, the fracture toughness properties of the as-forged specimens decreased with increasing yield strength. This trend is shown in Fig. 3. The screw press forging process at 816°C had the highest yield strength (495 MPa) and lowest fracture toughness values (1340 kJ/m²) of the as-forged steels. Table 3 and Figure 4 shows the average fracture toughness values as a function of decay helium content for the different forging processes and at the two forging temperatures. Tritium and decay helium caused fracture toughness values to be reduced by 80-90% compared to the as-forged condition for all of the forging processes, although the lowest average value was still greater than 150 kJ/m².

DISCUSSION

One of the objectives of this study was to measure the fracture toughness properties of Type 304L stainless steel as a function of forging temperature and forging deformation rate after tritium precharging and aging. Clear trends with these processing characteristics, however, did not emerge from these results. The higher forging temperature generally resulted in greater fracture toughness, although this was not always the case and likely represents scatter in the measurements. There is also no clear trend with forging rate for any of the measured conditions. Similarly, the role of annealing prior to the final stage of forging is inconclusive. Therefore, we consider the values in Table 3 collectively (i.e, independent of the processing characteristics): the average fracture toughness (J_Q) of the as-forged material is $> 1900 \text{ kJ/m}^2$ with a standard deviation of about 300 kJ/m². The material aged for one year (200 appm He) displayed a fracture toughness of $\sim 260\pm60 \text{ kJ/m}^2$, while the material aged for five years is $\sim 200\pm30 \text{ kJ/m}^2$.

Tritium-precharging produced a significant reduction in the fracture toughness values for all of the materials. The magnitude of the observed reduction is consistent with earlier studies on tritium effects on toughness [4-8]. There appears to be a steep decrease in fracture resitance with moderate amounts of helium content, although the baseline fracture resistance with tritium in the absence of heluim is not known. Comparison with published results on hydrogen-precharged forged Type 304L stainless steel suggests that the contribution of decay helium to toughness degradation is relatively small in relationship to the contribution of the hydrogen isotope. For example, fracture toughness of similarly forged Type 304L is reported [11] to be nominally in the range of 200-300 kJ/m². (with about twice as much precharged hydrogen based on solubility predicitons from Ref. [12] and with much larger specimens). In this study, the tritium content is decreasing as the helium content is increasing and the change in tritium content after one year and five years of aging was not measured. Nevertheless, extrapolation of the modest slope of the J_0 values at high helium content to zero helium content results in about 300 kJ/m², consistent with the forged and hydrogen-precharged Type 304L from Ref [11] and suggesting that the decrease of J_{Ω} can be attributed primarily to tritium.

CONCLUSIONS

Steels forged using the screw press forging process at 816°C had the highest yield strength (495 MPa) and lowest fracture toughness values (1340 kJ/m²).

The fracture toughness values of the mechanical press, hydraulic press, and HERF processes were very high and averaged more than 1900 kJ/m^2).

Tritium exposures reduced the fracture toughness by 80-90% from the asforged condition. The majority of this decrease can be attributed to the effect of hydrogen isotopes in the absence of decay helium.

Forging strain rate and temperature had no clear effect on fracture toughness properties after tritium exposures. Colletively for all conditions, J_Q values averaged ~270 kJ/m² and 200 kJ/m² for 200 and 600 appm decay helium respectively.

ACKNOWLEDGEMENTS

Savannah River National Laboratory is operated by Savannah River Nuclear Solutions and for the U.S. Department of Energy under contract number DE-AC09-08SR22470. Sandia National Laboratories is a multi-mission laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

REFERENCES

- G. R. Caskey, Jr., "Hydrogen Effects in Stainless Steels", Hydrogen Degradation of Ferrous Alloys, ed. J. P. Hirth, R. W. Oriani, and M. Smialowski, eds., (Park Ridge, NJ: Noyes Publication, 1985), p. 822.
- [2] S. L. Robinson, "The Effects of Tritium on The Flow and Fracture of Austenitic Stainless Steels", Hydrogen Effects on Material Behavior, A. W. Thompson and N. R. Moody, eds. (Warrendale, PA: TMS 1989) p. 433.
- [3] S. L. Robinson and G. J. Thomas, "Accelerated Fracture due to Tritium and Helium in 21-6-9 Stainless Steel", Metall Trans, 22A (1991), 879-885.
- [4] M.J. Morgan and M.H. Tosten, "Tritium and Decay Helium Effects on the Fracture Toughness Properties of Types 316L, 304L, and 21Cr-6Ni-9Mn Stainless Steels", Hydrogen Effects in Materials, A. W. Thompson and N. R. Moody, eds. (Warrendale, PA: TMS, 1996) p. 873.
- [5] M.J. Morgan, "Hydrogen Effects on the Fracture Toughness Properties of Forged Stainless Steels", 2008 ASME Pressure Vessels and Piping Division Conference, July 27-31, 2008, Chicago, Illinois USA.
- [6] M,J. Morgan, "Tritium Aging Effects on the Fracture Toughness Properties of Forged Stainless Steels", Proceedings of the Conference on Materials Innovations in an Emerging Hydrogen Economy, February 24-27, 2008, Cocoa Beach, Florida.
- [7] M. J. Morgan and M. H. Tosten, "Microstructure and Yield Strength Effects on Hydrogen and Tritium Induced Cracking in HERF Stainless Steel", Hydrogen Effects on Material Behavior, N. R. Moody and A. W. Thompson, eds. (Warrendale, PA: TMS, 1990) 447-457.
- [8] M. J. Morgan and M. H. Tosten, "Tritium and Decay Helium Effects on Cracking Thresholds and Velocities in Stainless Steels", Fusion Technol, 39, (2001) 590-595.
- [9] N.T. Switzner, C.J. Van Tyne, and M.C. Mataya, "Effect of Forging Strain Rate and Deformation Temperature on the Mechanical Properties of Warm-Worked 304L Stainless Steel", J Mater Processing Technol 210 (2010) 998-1007.
- [10] N.T. Switzner, T. Neidt, J. Hollenbeck, J. Knutson, W. Everhart, R. Hanlin, R. Bergen, D. K. Balch, C. San Marchi, "Hydrogen-Assisted Fracture in Forged Type 304L Austenitic Stainless Steel", Hydrogen-Materials Interactions, B.P. Somerday, P. Sofronis eds. (New York: ASME 2014) p. 273.
- [11] H. Jackson, C. San Marchi, D. Balch, B. Somerday, J. Michael, "Effects of low temperature hydrogen-assisted crack growth in forged 304L austenitic stainless steel", Metall Mater Trans 47A (2016) 4334-4350.
- [12] C. San Marchi, B.P. Somerday, and S. L. Robinson, "Permeability, solubility and diffusivity of hydrogen isotopes in stainless steels at high gas pressures", Intern J Hydrogen Energy 32 (2007) 100-116.

Table 1. Composition of Type 304L austenitic stainless steel used in this study.

Fe	Cr	Ni	Mn	Si	С	Ν	S	Р
Bal	19.48	10.69	1.63	0.52	0.029	0.03	0.0064	0.028

Table 2. Characteristics of forging processes, and resulting tensile strength properties of forged material from Ref. [9].

Process	Approximate Strain Rate	Forging Temperature	YS	UTS
	(s ⁻¹)	°C	(MPa)	(MPa)
Hydraulic	1	816	458	641
Mechanical	5	816	476	649
Screw	10	816	495	656
HERF	100	816	470	651
Prior Anneal + Screw	10	816	483	642
Prior Anneal + HERF	100	816	458	639
Hydraulic	1	871	412	617
Mechanical	5	871	436	628
Screw	10	871	461	631
HERF	100	871	444	637
Prior Anneal + Screw	10	871	461	631
Prior Anneal + HERF	100	871	433	629

Table 3. Average fracture toughness values of the as-forged and tritium-aged materials.

Fracture Toughness Values, J_Q (kJ/m²)

Process	Forging Temperature (°C)	As- Forged	Decay Helium Content 200 appm (est.)	Decay Helium Content 600 appm (est.)
Hydraulic	816	1915	340	153
Mechanical	816	1940	287	243
Screw	816	1340	309	182
HERF	816	1908	158	209
Prior Anneal + Screw	816	*	233	196
Prior Anneal + HERF	816	2129	216	*
Hydraulic	871	2490	*	229
Mechanical	871	1850	329	252
Screw	871	1880	208	215
HERF	871	2094	307	178
Prior Anneal + Screw	871	*	215	*
Prior Anneal + HERF	871	2031	235	*
*Not measured				



Figure 1. Shape and dimensions of fracture-toughness sample. Dimensions shown are in millimeters.







(b)

Figure 2. J-R behavior for screw press Type 304L austenitic stainless steel in the as-forged and tritium-precharged conditions for two forging temperatures: and tritium-precharged Type 304L austenitic stainless steel for two forging temperatures: (a) 816°C; and (b) 871°C. Similar behavior was observed for mechanical press, hydraulic press, and high-energy-rage forgings.



Figure 3. Fracture toughness values as a function of yield strength for all specimens before and after tritium precharging and aging.



Figure 4. Average fracture toughness values as a function of decay helium content for each of the four forging processes and two forging temperatures: (a) 816°C; and (b) 871°C. Zero helium content represents the as-forged condition.