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EXPERIMENTAL DETERMINATION OF THE EFFECT OF HELIUM ON THE FRACTURE TOUGHNESS OF STEEL

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A fundamental issue with the use of structural steels for fusion applications is the effect of helium on mechanical properties. This concern has been difficult to address due to the limited neutron energies, hence limited helium production, of the fission reactor facilities used to simulate the effects of irradiation on properties such as fracture toughness. This paper compares results from identical pre-cracked DCT fracture toughness samples irradiated using spallation and fission neutrons at ~60-90°C. Materials studied were 304L and 316L stainless steel. The spallation neutron irradiated specimens were irradiated over a dose range up to ~ 10 dpa with a helium and hydrogen-to-dpa ratio of approximately 60 and 400 appm/dpa, respectively. Fracture toughness was seen to rapidly decreased from a value of ~250 MPa-m$^{1/2}$ to ~ 200 MPa-m$^{1/2}$ by the 1 dpa level. Following fission irradiation in the 1-1.5 dpa range, fracture toughness results were indistinguishable from those irradiated in the spallation neutron. The calculated helium and hydrogen concentration for the fission neutron irradiated materials was ~ 1 appm and ~30 appm, respectively. It is concluded that, within measurement uncertainty, increasing the helium and hydrogen concentration by more than an order of magnitude had little influence on the fracture toughness of 304L and 316L stainless steel irradiated to similar displacement doses of ~ 1 dpa in the temperature range of 60-90°C.

Introduction

Austenitic stainless steels are the primary structural materials for the in-vessel components of ITER. Specifically, the 316LN-IG steel will be used as the structural materials for the blanket system which will be cooled at ~140°C. Selection of this grade of steel was based on more than twenty years of research and operating experience in light water and fast breeder reactors. [1] A persistent question regarding the use of steels in fusion systems is the effect of transmutation gasses on microstructure and mechanical properties. On a dpa normalized basis, the helium and hydrogen produced by fusion neutrons will be on the order of 15 appm/dpa He and 60 appm/dpa H.[2] As the data-base used to study the irradiation effects on stainless steels has been primarily generated using fission reactors, which have a much lower gas production, the question is raised as to whether there is an intrinsic helium or hydrogen effect on the mechanical properties.

While there is likely to be little difference in the surviving defects, and thus the mechanical properties on a dpa-normalized basis when comparing fission and fusion spectrums, the effect of high levels of helium on the mechanical properties of neutron-irradiated steels has long been debated and remains unanswered. Fracture toughness models based on matrix hardening predict a negligible effect of helium on fracture toughness. However, recent experiments on ferritic/martensitic steels based on nickel doping, boron doping, and cyclotron injection appear to indicate a helium effect on low temperature embrittlement. A common concern with such experiments involves the possible effects doping has on the microstructure and fracture properties. Likewise, the helium injection method suffers from difficulty in interpretation of data from severely size-constrained specimens. The transmutation question has become a focal point of the international fusion materials community leading to a call for a dedicated facility (IFMIF) to study the effect

The Accelerator Production of Tritium (APT) project undertook to generate design data for various target/blanket materials using the Los Alamos Neutron Scattering Center (LANSCE) to simulate high-energy neutron damage. This program is now complete and has provided high quality data on the mechanical properties of 304L, 316L, Modified 9Cr-1Mo, various aluminum alloys and Alloy 718.[3,4] The irradiation conditions were from 0.1-10 dpa, irradiation temperature ranged from 50-160°C, and helium and hydrogen-to-dpa ratios was approximately 60 and 400, respectively. The existence of this data, generated from materials with even higher helium and hydrogen-to-damage ratios than would occur in fusion, presented a unique opportunity to gain insight into the questions regarding the effect of transmutation gas on mechanical properties of structural materials. This paper compares fracture toughness results on identical

304L and 316L steels, identically tested, irradiated to similar dose levels and temperatures in the LANSCE facility and the High Flux Isotope Reactor (HFIR,) thereby giving a single variable comparison of the effect of transmutation gasses on the fracture of irradiated austenitic steel.

Experimental

The 304L and 316L specimens used for both LANSCE and HFIR irradiation were EDM'd into disc compact tension (DCT) specimens from self-similar heats in the as-received (annealed) condition. A schematic drawing of the DCT specimen is given in Figure 1. Table 1 gives their chemical compositions of the alloys and optical images of their polished microstructures. Specimens were fatigue precracked before irradiation to a ~0.5 ratio of the crack length to specimen width, and then side-grooved by 20% of their thickness (10% from each side.) The J-R-curve toughness tests were carried out in general accordance with the ASTM Standard E 1820-99. Specimens for LANSCE irradiation were both 2 and 4 mm in thickness, while those irradiated in HFIR were only 2 mm. Results from the LANSCE irradiation indicate that constraint, or other effects, did not significantly effect the data yielding similar results for both 2 and 4 mm thick DCT specimens. Detailed information on test method is provided by Sokolov.[3] A discussion of the validity of fracture toughness results and the methodology for obtaining fracture toughness from the DCT specimens is found elsewhere. [3,5]

Spallation neutrons were produced by the LANSCE facility utilizing an 800 MeV, 1 mA Gaussian proton beam interacting with a tungsten target. The fluence was determined by evaluation of activation foils. Details of dose (and dpa) determination is discussed elsewhere by Maloy. [3,4] Irradiation temperature was estimated by a combination of calculation and active thermocouple measurement. [6] The fission neutron irradiation was carried out in the target region of the HFIR. The calculation of dpa follows the HFIR target-specific calculations of Greenwood, et al. [7] which gives a correlation of 0.734 dpa/10^{25} n/m^2(E>0.1 MeV.) The irradiation was carried out in contact with the HFIR coolant water of ~ 50°C. Considering the gamma heating (~ 40 W/g) in this position, the center temperature of the DCT sample would be ~ 60°C.

An example of the raw data for LANSCE irradiated 304L is given in the Figure 2. Data on displacement was obtained using an outboard clip gage utilizing razor-blades seated in grooves machined on the outer edge of the specimen above and below the loading holes (figure 1.) Figure 2 data exemplifies the load-unload compliance method indicating a material that is failing by controlled crack extension. All samples in this study exhibited such stable crack extension behavior.

Results and Discussion

The temperature-dependent irradiation-induced defect structure in austenitic stainless steels is rather complex, though well understood. At lease six different planar and three-dimensional interstitial and vacancy defects have been analyzed and at least eight radiation-induced, or radiation-modified phases have been identified. [8] The irradiation temperature of this study corresponds to a regime of onset of vacancy motion with limited ability of vacancy clusters formed within cascades to become thermally unstable. In this temperature regime, the fission neutron damage is characterized by 1-2 nm “black-spot” defect clusters. It has been shown that in the temperature regime of 100-200°C the character of these defect structures is the stacking fault tetrahedral (SFT’s,) with ~ 20% of density of the defects being small interstitial loops. [9] As the irradiation temperature is increased, the fraction of SFT’s decreases and at ~300°C austenitics begin to form small cavities. A limited amount of microstructural observation of the LANSCE irradiated alloys has been conducted, with the general observation that results are in qualitative agreement with published information on fission reactor irradiated materials. As example, Sencer[10] observed a density of black spot defect clusters, which decrease in density with irradiation temperature, in general agreement with the fission reactor irradiated austenitic stainless steel literature.

In the low temperature regime of this study, the yield strength of austenitic stainless steel is known to rapidly increase and saturate by ~ 1 dpa. [11] This is attributable to the rapid build-up of clusters, Frank loops, and SFT’s. Figure 3 gives the dose dependence of fracture toughness and transmutation gas concentration for LANSCE irradiated specimens. The fracture toughness is seen to decrease significantly for both austenitic alloys at the 0.1 dpa level. For both alloys, the toughness decreases from ~ 250 to ~ 175 MPa-m$^{1/2}$. This degradation in K_{1c} is directly attributable to the rapidly increased yield strength, which has been measured for tensile specimens also irradiated under the APT program. [4] The APT results agree qualitatively with the fission reactor data, exhibiting a rapid rise in yield strength which saturates by 1 dpa. However, as pointed out by Maloy,[4] who compared the APT tensile results with fission reactor irradiated 300 stainless steels irradiated below 100 (though not identical materials to those of the APT study,) there appears to be a difference in the strain-to-necking (stn) for the two irradiation sources. Specifically, both 304L and 316L LANSCE irradiated specimens degraded to essentially zero stn by ~ 5 dpa, while the fission reactor irradiated material retain >20% stn.

As the dose for the LANSCE irradiated austenitics increases (figure 3,) the fracture toughness appears to approach saturation, with toughness remaining > 50 MPa-m$^{1/2}$. The calculated helium and hydrogen concentration is given in figure 3. For spallation irradiation, the transmutation of gas occurs in a nearly linear fashion. In this case, helium and hydrogen-to-dpa ratios of approximately 60 and 400, respectively, were achieved. On a dpa normalized basis, this is significantly higher than would be transmuted in a fusion neutron spectrum (~15 appm/dpa He and 60 appm/dpa H.[2]) It is interesting to note that the ~9.3 dpa irradiation (figure 3,) the material contained ~ 700 appm He and nearly 6000 H, and still remaining tough. An important side-note is that Oliver [12] has experimentally determined that these high levels of transmutation gas remained in the samples following irradiation.

Figure 4 gives the comparison of the LANSCE and HFIR irradiated 316L and 304L steels. The helium and hydrogen concentration for the HFIR irradiated materials was ~ 1 appm and ~30 appm, respectively.[2] As seen in the figure, the dose range for the HFIR irradiation was rather limited, ranging from ~ 1-1.5 dpa. In this dose range, the temperature of the HFIR irradiated materials (~ 60°C) compares well with the LANSCE irradiated materials (50-80°C.) From the data, it is seen that within the statistical scatter, the fracture toughness of the HFIR irradiated 316L steel is consistent with the LANSCE irradiated materials in the range of 188-215 MPa-$m^{1/2}$. It should be noted that there was a very weak temperature dependence of fracture toughness for these materials.[3] On figure 4, values of fracture toughness taken at 25 and 50°C can be considered equivalent.

The fracture toughness of the 304L steel (figure 4) is showing a more interesting behavior. While the LANSCE irradiated materials shows the significant drop at the 0.1 dpa to ~175 MPa-$m^{1/2}$ followed by a shallow decrease to about 150 MPa-$m^{1/2}$, the HFIR irradiated materials appear to be somewhat tougher at the ~ 1-1.5 dpa dose level. Comparing the data on figure 4 it is seen that the HFIR irradiated 304L has a toughness of ~ 220 MPa-$m^{1/2}$ compared to an extrapolated value of ~ 160 MPa-$m^{1/2}$ for the LANSCE data. As there are four quality measurements essentially showing the same toughness over this dose range, it is difficult to ascribe the differences to statistical variation.

Conclusion

1) Alloy 316L steel irradiated with fission neutrons at ~ 60°C, containing ~ 1 appm He and ~ 30 appm H, yields similar as-irradiated toughness (~200 MPa-$m^{1/2}$) at 1-1.5 dpa, as spallation neutron irradiated 316L containing ~ 60 appm He and 400 appm H. Within the limits in dose and temperature of this study it appears that helium and hydrogen have little effect on the fracture toughness of 316L.

2) Under the same irradiation comparison as the 316L, Alloy 304L appear to have a slightly higher toughness for the HFIR irradiated, lower helium and hydrogen content, samples. Specifically, the HFIR irradiated 304L was tougher by ~ 60 MPa-$m^{1/2}$ at the ~ 1 dpa level. Further investigation will be required to determine possible explanations for the difference between the two alloys.

Acknowledgement

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Figure 1 : Schematic of Disc Compact Tension Specimen

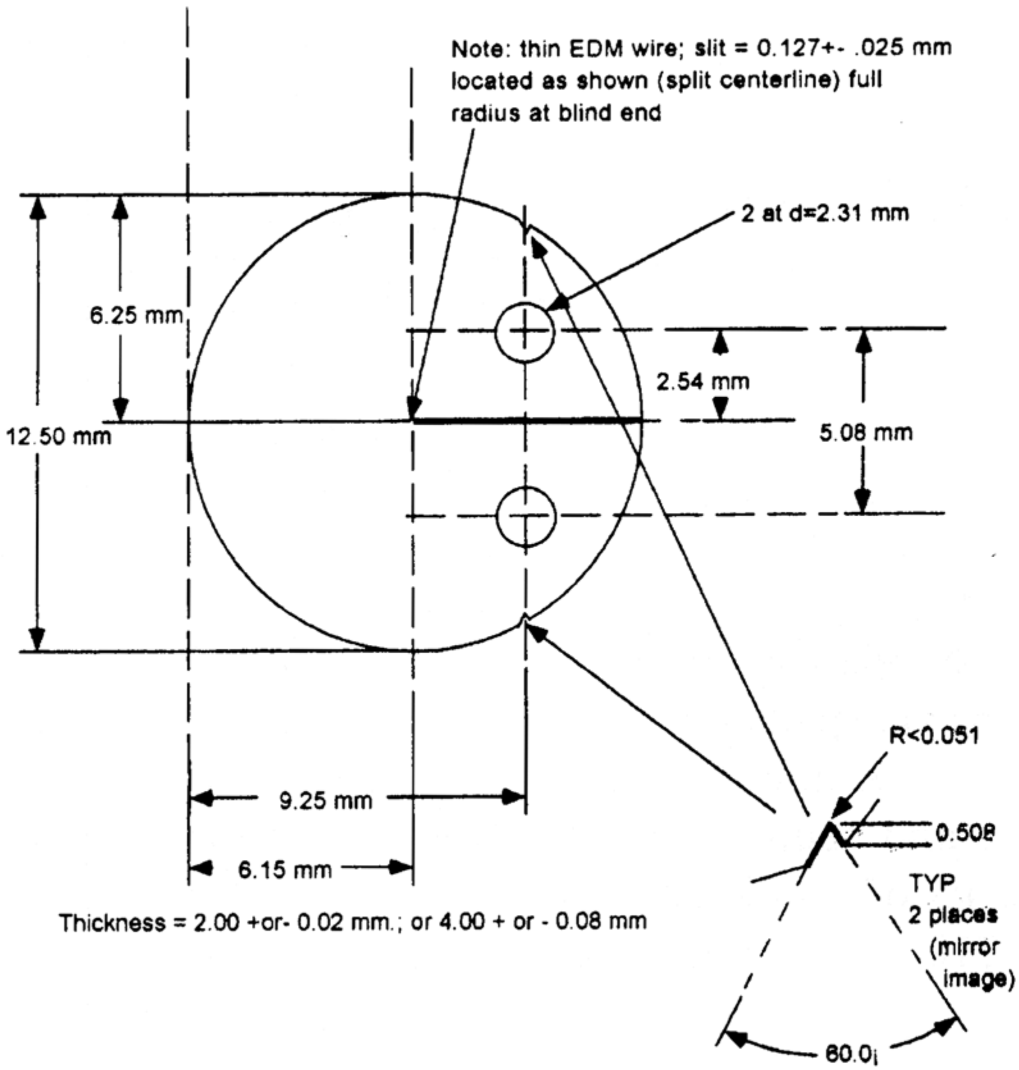


Figure 2 : Load-Displacement Curve for LANSCE Irradiated 304L

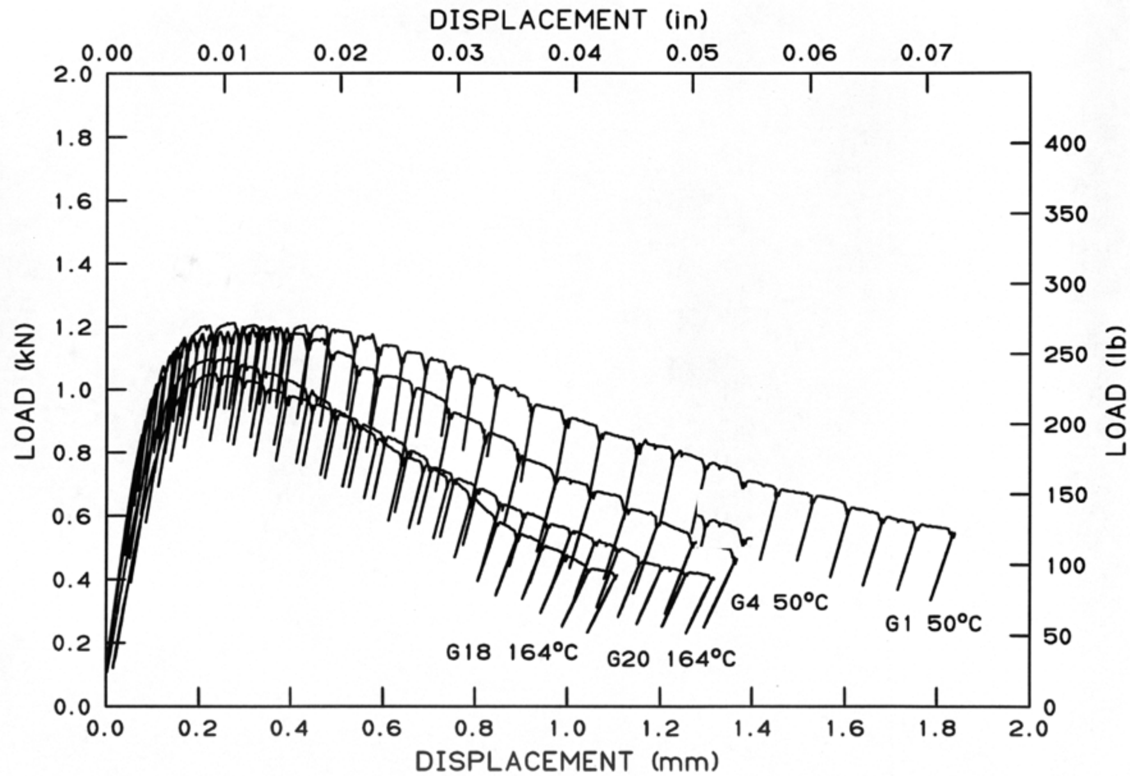


Figure 3 : Dose Dependence of Fracture Toughness and Transmutation Gas Concentration for LANSCE Irradiated Specimens

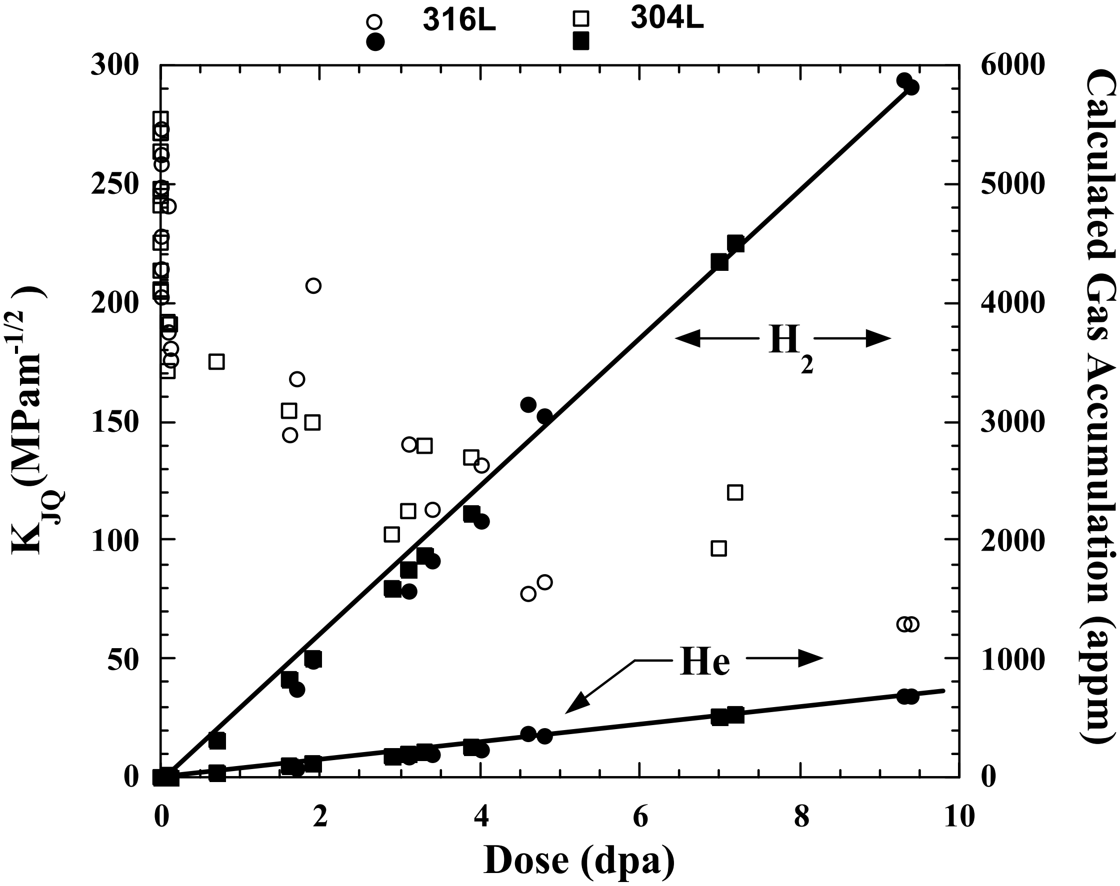
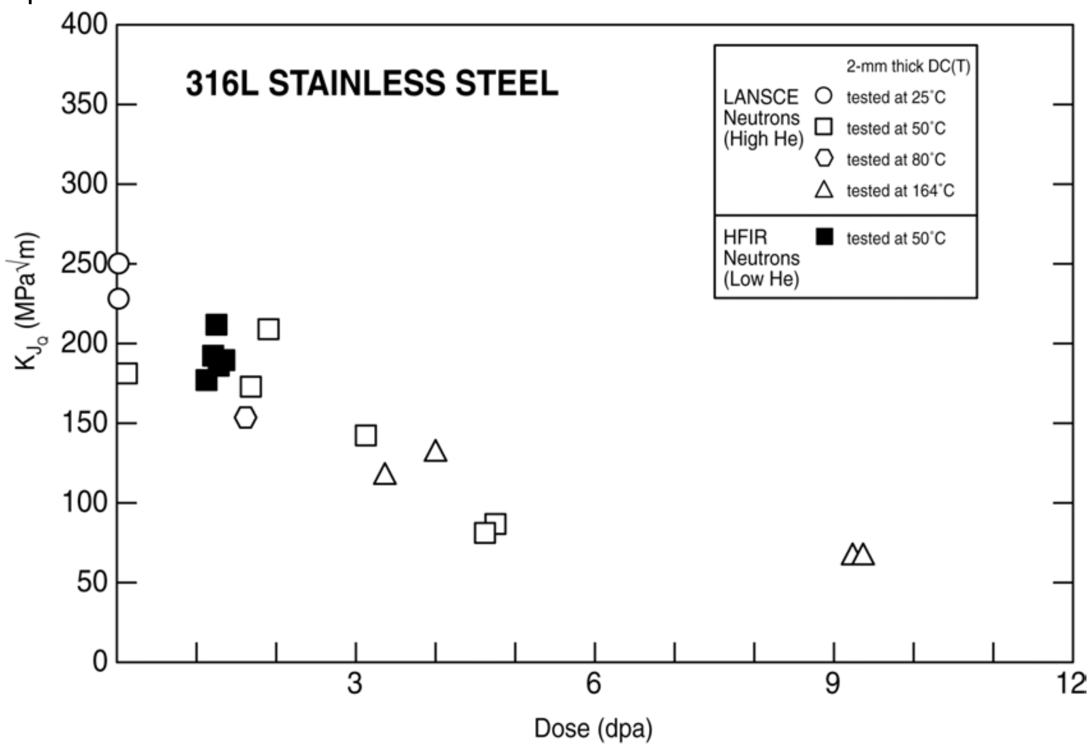


Figure 4 : Fracture Toughness Comparison of HFIR and LANSCE Irradiated Specimen



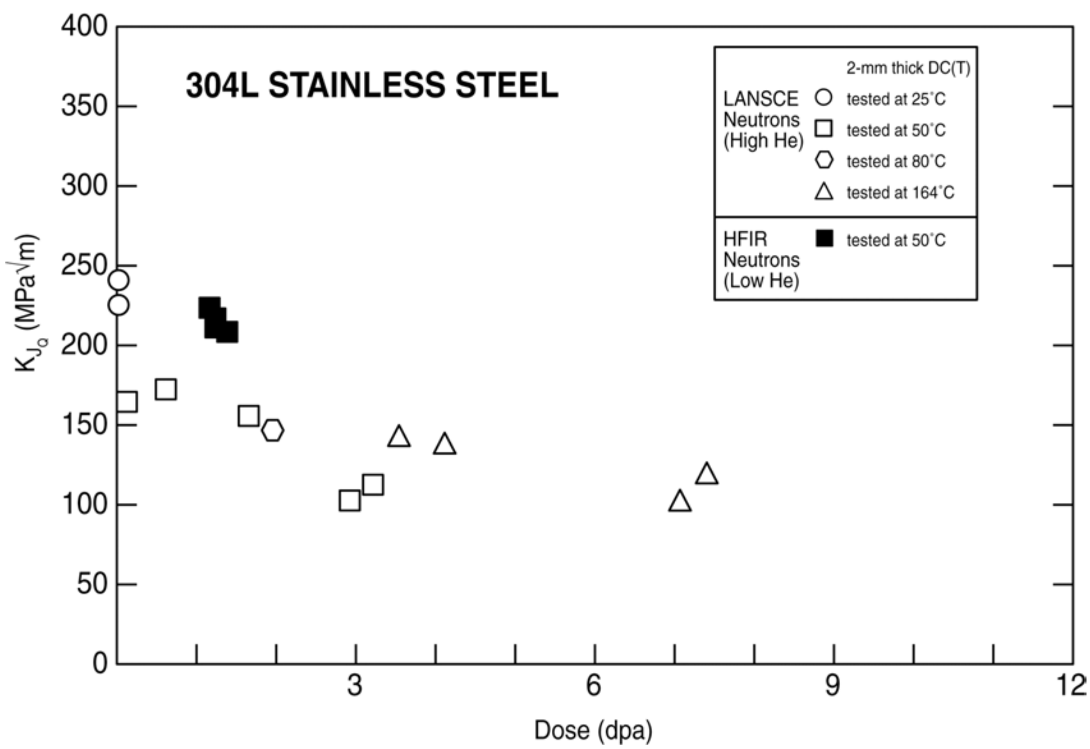
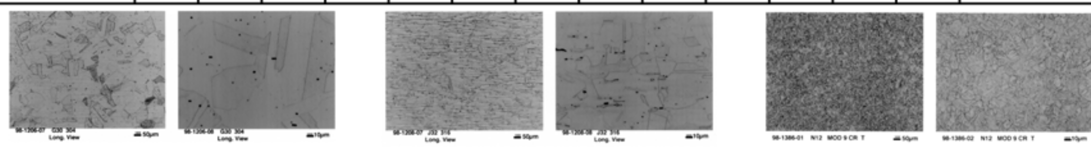


Table 1 : Materials irradiated in LANSCE and HFIR.

Material	Lot	A1	C	Cr	Cu	Fe	Mn	Mo	Ni	P	S	Si	Ti	Others
304L, 2, 4	K681		0.01	18.15	0.23	Bal	1.80	0.18	8.35	0.025	0.010	0.43		Co-0.17; N-0.085
316L, 2, 4	D306		0.01	17.33	0.18	Bal	1.61	2.09	10.62	0.024	0.019	0.43		Co-0.21; N-0.060
Mod 9Cr-1Mo	10148	.002	.089	9.24	0.08	Bal	0.47	0.96	0.16	0.021	0.006	0.28	0.002	V-0.21; Nb-0.054; Co-0.019; N-0.035; O-000.008



304L Stainless **316L Stainless** **9Cr-1Mo**

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