ANALYSIS OF A CLAD TUNGSTEN TARGET AFTER IRRADIATION IN AN 800 MeV PROTON BEAM

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ABSTRACT

For the Accelerator Production of Tritium (APT) and the Accelerator Driven Transmutation Facility (ADTF), tungsten is being proposed as a target material to produce neutrons. Previous work showed that the mechanical properties of tungsten are degraded from irradiation in a neutron flux but little work had been performed on the irradiation of tungsten in a high energy proton beam. In this study, tungsten rods were irradiated at the 800 MeV Los Alamos Neutron Science Center (LANSCE) proton accelerator for six months. To avoid corrosion during irradiation, the rods were clad with a thin (0.25 mm thick) 304L or (0.125 mm thick) annealed alloy 718 tubing. After irradiation to a maximum dose in the tungsten of 23.3 dpa, the clad rods were opened in the hot cells and the tungsten was removed. The tungsten was then sliced into short compression specimens (~3mm long). Hardness tests and compression tests were performed on the tungsten rods to assess the effect of irradiation on their mechanical properties. Results show an increase in hardness with dose and irradiation temperature and an increase in yield stress with dose.

1. INTRODUCTION

Tungsten is being considered for use as a primary or backup neutron source in many spallation neutron source applications such as the APT, ADTF, the Spallation Neutron Source (SNS), KENS (the spallation neutron source at the High Energy Accelerator Research Organization, KEK) and the Accelerator Transmutation of Waste (ATW) projects. For such applications its irradiation temperature is close to the brittle-to-ductile transition temperature for tungsten before irradiation which ranges from 65-700ºC depending on the impurity content, grain size and heat treatment of the tungsten [1], [2], [3]. Therefore, tungsten is quite notch sensitive in this temperature regime making it difficult to measure its tensile properties. Very often, the tungsten specimens break in the elastic region before reaching yield [4], [5]. Therefore to avoid brittle facture, the mechanical properties of tungsten have been studied in compression after irradiation in a proton beam.

2. BACKGROUND

The effects of irradiation on tungsten have been studied previously but have mainly centered on the recovery of defects in irradiated tungsten with a few papers relating to the effect of irradiation on the mechanical properties of tungsten. The irradiation temperature of the tungsten in this paper is between 50 and 270ºC. These temperatures are in the stage III recovery range for tungsten. Much debate has centered on the defects responsible for recovery in stage III. Kim and Galligan[6], present strong arguments that the interstitials must be the mobile defects responsible for recovery during this stage as single vacancies are always observed after stage III recovery and the measured activation energy, 1.7 eV, is too low for vacancy migration.

A few papers have been written on the mechanical properties of tungsten after irradiation, [7], [2], [5], [3]. In these studies, the mechanical properties were either measured in bending, in tension or through hardness measurements. When the properties were measured in bending or tension (at 300ºC or below), the specimens broke in the elastic regime or after very low elongations at 200ºC (less than 1% uniform elongation[2]). Thus it is quite difficult to compare to these results because true yield was not measured. In one report the hardness was measured after irradiation in a proton beam [7]. These results showed an increase in hardness from 489 to 563-583 kg/mm^2 after irradiation to a dose of 3.7x10^{20} protons (~2.4 dpa). The calculated irradiation temperature was 120-300ºC.

In this paper the mechanical properties of tungsten will be presented after irradiation in an 800 MeV, 1mA proton beam to a maximum dose of 23 dpa. The properties were measured by means of compression testing and hardness testing.

3. EXPERIMENTAL

99.95% purity tungsten was obtained from Schwarzkopf Technologies Corporation[1] in the form of ~3 mm diameter rods. Two different heats of tungsten were irradiated. One was 2.6 mm in diameter and a second was 3.2 mm in diameter. These rods were slip clad with either 0.25 mm thick 304L tubing (for the 2.6 mm diameter rods) or 0.125 mm thick alloy 718 tubing (for the 3.2 mm diameter rods) and backfilled with helium. Bundles containing 19 rods each were held in tubes and cooled with flowing water [8]. The 2.6 mm diameter rods were irradiated for six months and the 3.2 mm diameter rods for two months with an 800 MeV, 1mA proton beam with a Gaussian distribution where two sigma= 3.2 cm. Each tungsten rod was 10 cm long allowing the accumulation of a range of doses on each rod from the center of the rod to the edge.

The fluence determination (see results in Table 1) for the irradiated samples was performed through analysis of an activation foil package that was irradiated in the center of each clad rod. The activation foil packages were TEM-sized disks punched from >99.98% pure sheet material of Al, Fe, Co, Ni, Cu, and Nb. After irradiation, the stacks were withdrawn and counted with gamma spectroscopy to quantify the isotopes produced. This provided several reactions with various cross sections and thresholds which were used to estimate the proton and neutron group fluxes. The production rates of the isotopes were calculated by taking into account the proton beam history and the measured activity. Proton and neutron flux estimates were calculated using the MCNPX code[9]. The input fluxes were then adjusted to match the measured isotope production rates using the STAYSL2 code [10]. The revised fluxes for protons and neutrons were then folded with He, H and dpa cross sections for the materials of interest. This firmly established the exposure parameters at the activation foil locations. The error associated with the fluxes and damage levels was estimated to be around 25%.

Irradiation temperatures in the clad tungsten rods were determined as a function of position along the rods using LAHET Code System[11] calculated local power densities as input. The 2.6 mm rods were located in an insert with only one other materials insert in the beam before it. So the peak power density was 2250 W/cc. The 3.2 mm rods were located in an insert with several inserts in the beam before it so the peak power density was only 1020 W/cc. For both inserts there was more than a factor of 10 difference in power density between the tungsten at beam centerline and at the end of the rods. Cooling water temperatures were calculated locally from measured input values. The cooling water (T_o) at the start of the bundle was 27.6ºC for the 2.6 mm rods and 34.8ºC for the 3.2 mm diameter rods.

Tungsten irradiation temperatures, T_{irr}, (see Table 1) were calculated at each location along the rod as follows. First the heat transfer coefficient was calculated for the water flowing in the space between the 19 rods in the tube. The temperature drop from clad surface to water (ΔT_{film}) was calculated by dividing the heat flux coming out of the clad by the heat transfer coefficient. The temperature difference through the clad (ΔT_{clad}) was determined by calculating the contributions from the heat flux coming into the clad from the tungsten and the power density in the clad itself. The temperature difference through the helium gap (ΔT_{gap}) was calculated as conduction of the entering heat flux from the tungsten rod through the helium gas. The temperature rise from the tungsten rod surface to rod centerline (ΔT_{rod}) was calculated using the tungsten power density with radial conduction through the rod. The peak tungsten temperature (T_{irr}) at each location along the rod is then:

$$T_{irr}=T_o+\Delta T_{film}+\Delta T_{clad}+\Delta T_{gap}+\Delta T_{rod}.$$

Compression specimens were prepared from one 2.6 mm diameter rod and one 3.2 mm diameter rod by slicing the rod with a slow speed diamond saw into ~3 mm long segments. Then the faces were ground parallel using 600 grit SiC paper. The exact diameter and length of each specimen was measured before testing. Then a dab of vacuum grease was applied to the ends of each specimen. Specimens were tested in compression at an initial strain rate of 10^{-3}/s using an Instron 5567 mechanical testing machine in a hot cell. Load/displacement curves were converted to stress/strain using the initial measurements

Table 1 Irradiation Conditions for Tungsten Specimens

Sample #	Dose (dpa)	Tirr (C)	Calculated H (appm)	Calculated He (appm)
W1-3	21.9	250	10300	1900
W1-5	17.6	190	8300	1500
W1-6	14.9	160	7000	1300
W1-7	2.8	50	1300	250
W1-8	3.2	50	1500	270
W1-9	3.7	50	1800	320
W1-10	4.6	60	2100	400
W1-12	4.0	160	1600	290
W1-13	3.8	160	1600	280
W1-16	2.8	120	1100	200
W1-17	0.6	60	200	40
W1-18	0.7	60	300	50
W1-19	0.9	60	400	70
W1-21	1.5	80	600	110
W1-22	23.3	270	11000	2020

[1] Schwarzkopf Technologies Corporation, 35 Jeffrey Ave., Holliston, MA 01746-2028

and compliance from the test frame was removed from each curve. Some specimens were mounted in epoxy and polished to 1 micron diamond paste. Then, diamond pyramid hardness tests were performed using a Leitz Metallograph with a 400g load.

4. Test Results

The stress/strain curves for the specimens irradiated at 120-270ºC are shown in Fig. 1 and the specimens irradiated at 50-80ºC in Fig. 2. Stress/strain curves for specimens irradiated at 50-80ºC exhibit a larger yield drop over those irradiated at 120-270ºC. Each test was stopped after accumulating ~20% plastic strain. A slightly higher yield stress was measured for the 0 dpa 2.6 mm specimen over the 0 dpa 3.2 mm specimen. Two tests in Fig. 1 began to show a decrease in load from splintering of the specimen during testing. All tests show an increase in yield stress with dose.

Photographs were taken of the sides of the specimens after testing. Cracking typical of that observed on almost all irradiated specimens is shown in Fig. 3b compared to a non-irradiated specimen in Fig. 3a. All irradiated specimens exhibited longitudinal cracking after testing except for one specimen (W1-7). This was the lowest dose 2.6 mm diameter specimen (2.8 dpa) and it is possible that microcracking occurred that was not visible with the low magnification (16X) optical microscope used for analysis.

The averages of five measured hardness values are plotted vs. dose in Fig. 4 (error in measurements is less than 2%). The hardness increases quickly after 0.8 dpa but only slightly increases for the same irradiation temperature out to 4 dpa. When the irradiation temperature is increased to 160ºC, the hardness increases at the same dose, 4 dpa and continues to increase with increasing dose out to 23 dpa.

5. Discussion

The effect of irradiation temperature on the mechanical property results is clearly shown when comparing stress/strain curves for specimens irradiated to a similar dose while varying the irradiation temperature (see Figure 5). Stress/strain curves are shown for two specimens irradiated to 2.8 dpa at temperatures of 120 and 50ºC and two specimens irradiated to 4.6 and 4.0 dpa at 60 and 160ºC, respectively. All specimens began yielding at ~1600 MPa but large yield drops are observed in both specimens irradiated at 50 and 60ºC while much smaller yield drops are observed for specimens irradiated at 120 and 165ºC. One possible explanation for this difference is that either larger interstitial clusters or helium bubbles are formed after irradiation at higher temperatures which cannot be swept away by dislocation motion while at lower temperatures a uniform distribution of small interstitial clusters are formed which can be swept away by dislocation motion during initial yielding. In future work, TEM analyses will be performed to investigate the irradiated microstructure.

Hardness results are compared to previous results by Sommer et al.[7] in Figure 4. A much higher hardness is reported after ~2 dpa of exposure. These results are also from irradiation at a higher temperature, 150-300ºC, which may explain the higher observed hardness. Our results show that for two specimens irradiated to 4 dpa, an increase in irradiation temperature from 50 to 160ºC results in an increase in hardness by 40 kg/mm^2.

Cracking observed on the sides of compression specimens after irradiation are an indication of a decrease in ductility in tension. Such a decrease in ductility has been observed in results from testing neutron and proton irradiated materials. Tungsten bend specimens irradiated in a 800 MeV proton beam to 2.4 dpa exhibit zero ductility (break in the elastic regime) at 150ºC[7]. In addition, neutron irradiated specimens ($1x10^{21} n/cm^2$) show zero ductility after irradiation and testing at 300ºC[5] and an increase in DBTT by 150ºC after irradiation at 385ºC to $9x10^{21} n/cm^2$ [3] and an increase in DBTT by 165ºC after irradiation to $9.5x10^{20} n/cm^2$ at 250ºC[12].

6. Conclusions

The effect of proton irradiation on the mechanical properties of tungsten has been measured through hardness and compression testing after irradiation in a proton beam to a maximum dose of 23 dpa. The results show the following:

1. The yield stress of tungsten increases by almost a factor of 2 after irradiation to 23 dpa.
2. Specimens irradiated at 50-80ºC exhibit a larger yield drop in the compression stress/strain curves and a higher hardness over those irradiated at 120-270ºC at the same dose.
3. Cracking is observed on the sides of compression specimens after testing suggesting a decrease in ductility after irradiation.

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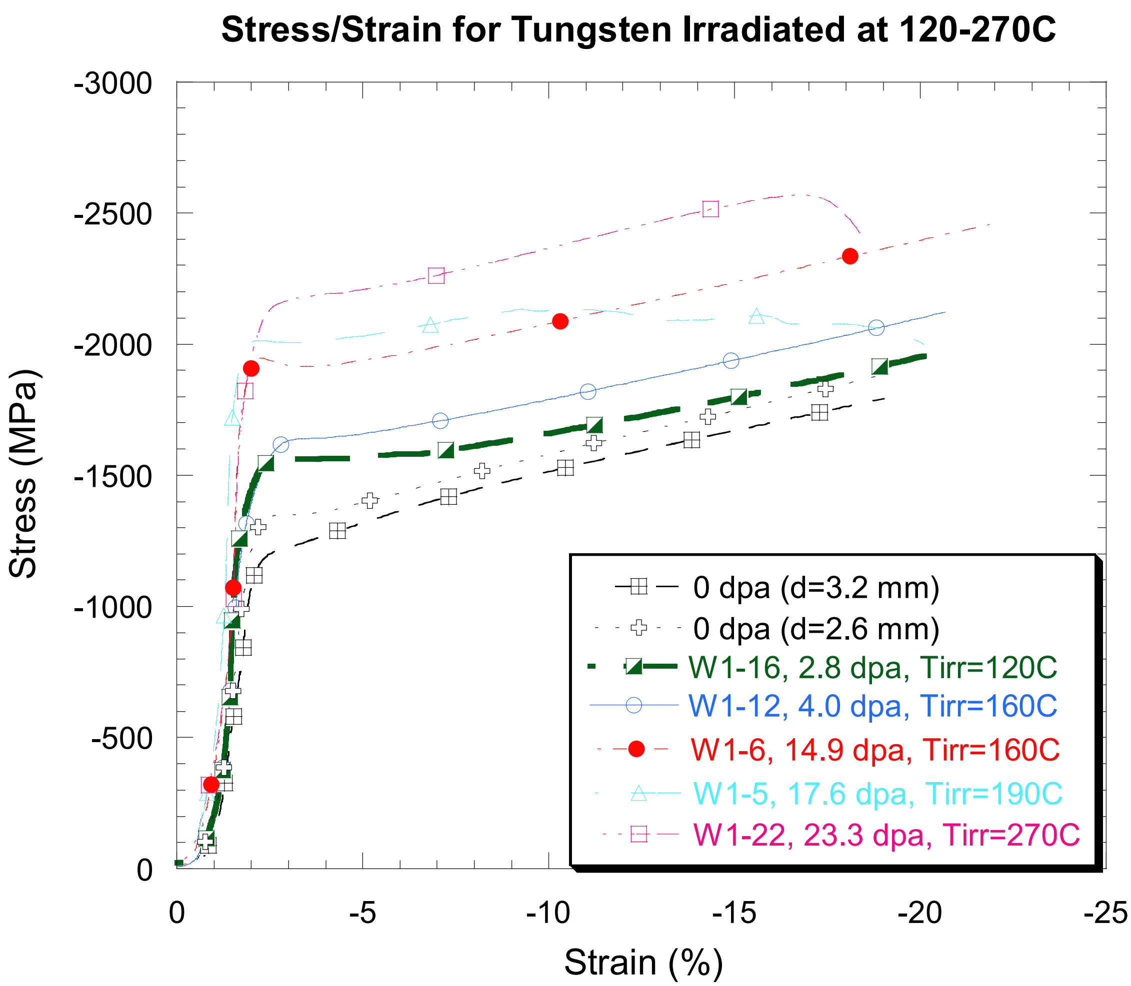


Figure 1 Graph showing stress/strain curves for tungsten tested in compression after irradiation in a proton beam to a maximum dose of 23 dpa at 120-270ºC.

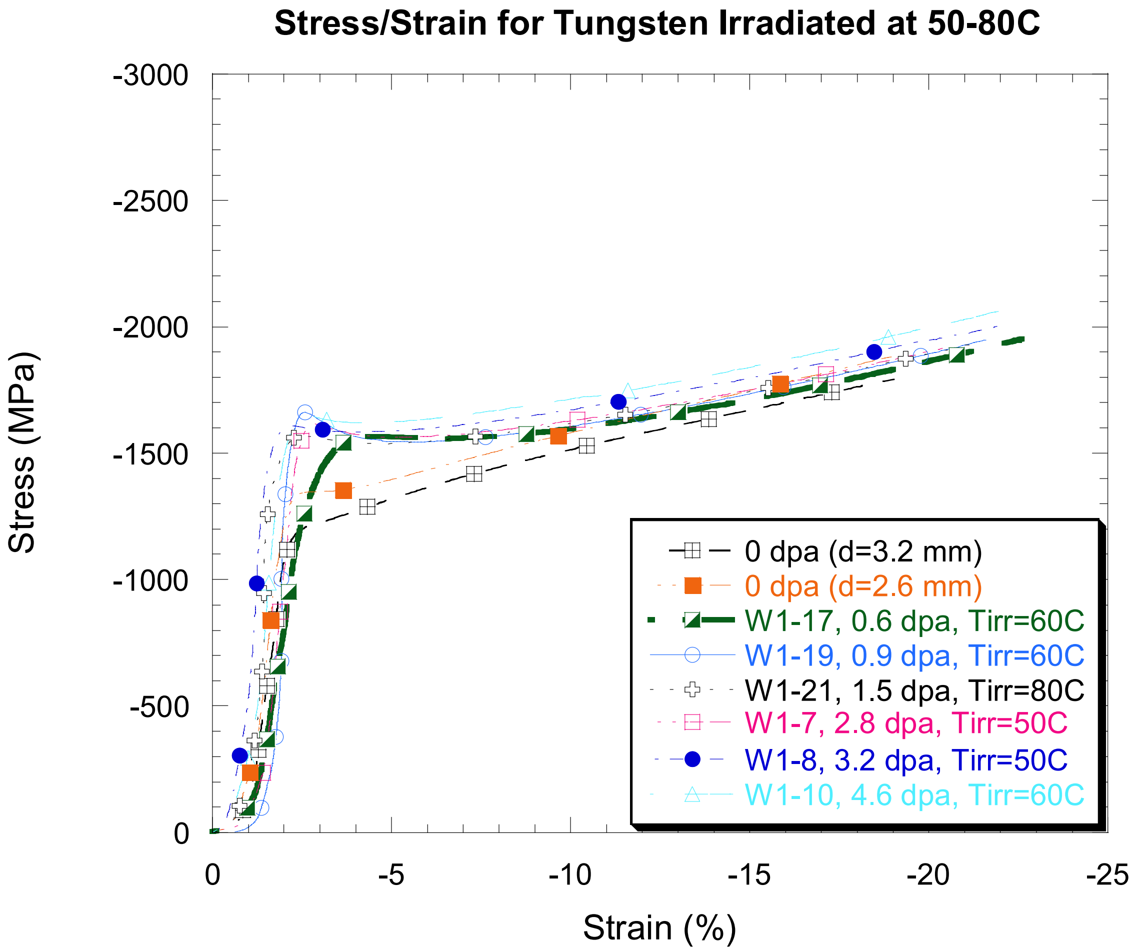
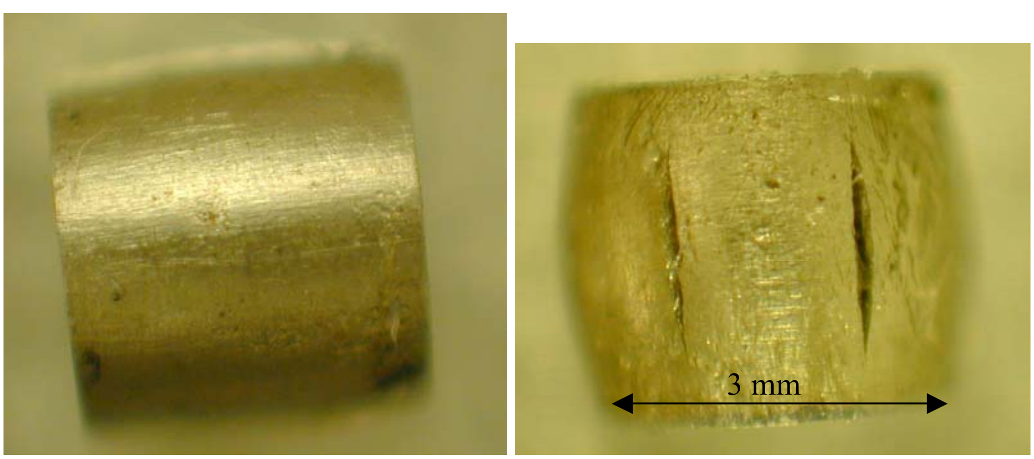


Figure 2 Graph showing stress/strain curves for tungsten tested in compression after irradiation in a proton beam to a maximum dose of 4.6 dpa at 50-80ºC.



A **B**

Figure 3 Optical micrographs of tungsten compression specimens after compression to ~20% strain before irradiation (A) and after irradiation to 23.3 dpa (B).

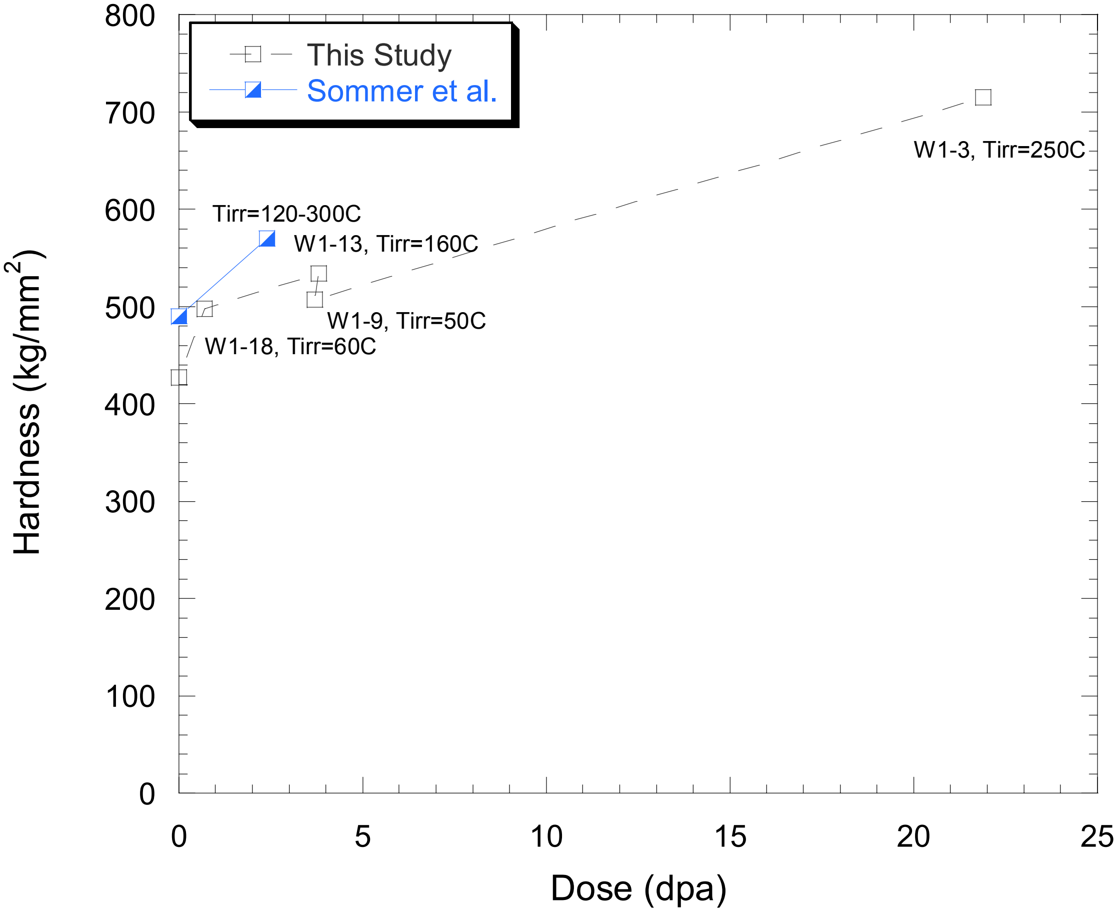


Figure 4 Graph plotting the change in hardness with dose in tungsten after irradiation in a proton beam at 50 to 300ºC.

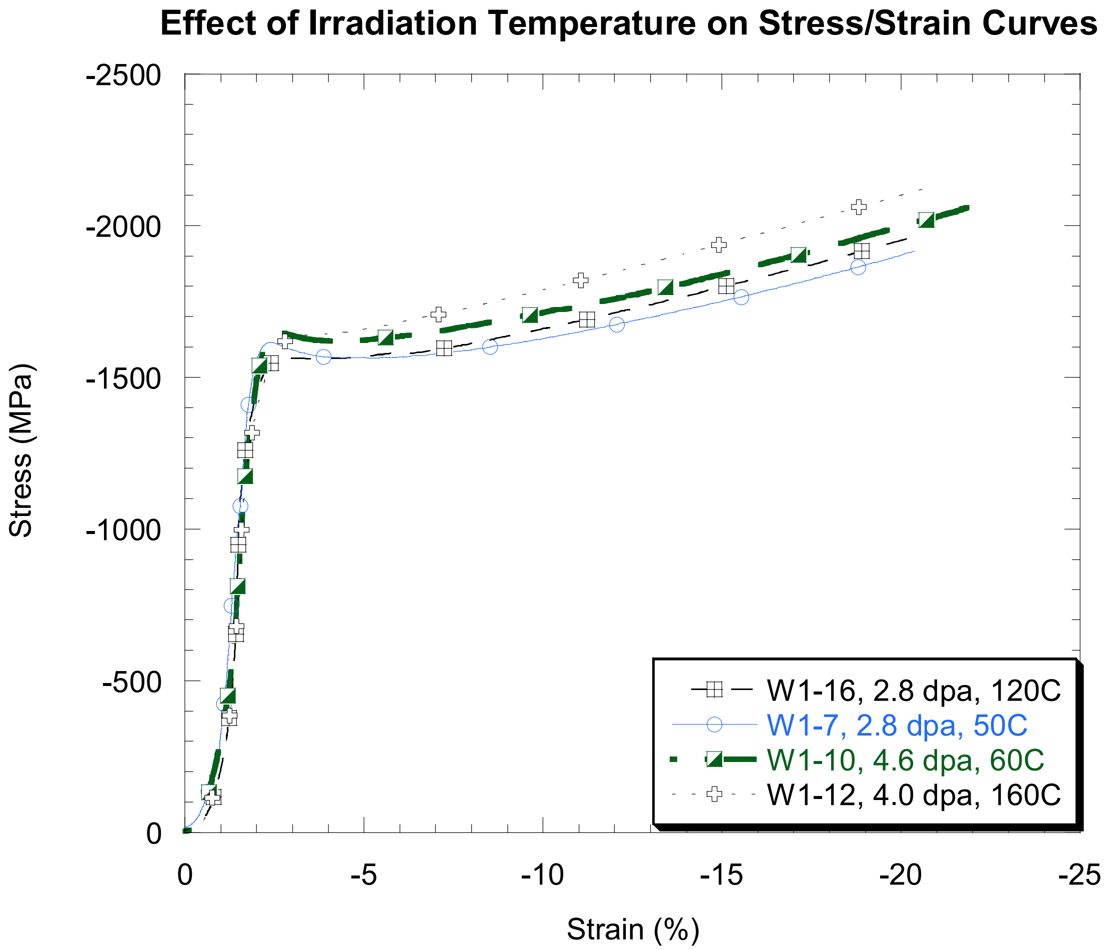


Figure 5 Graph comparing stress/strain curves for tungsten measured in compression after irradiation in a proton beam to 2.8 dpa and 4.0-4.6 dpa at irradiation temperatures from 50 to 160ºC.