

EFFECTS OF IRRADIATION AT LOW TEMPERATURE ON V-4Cr-4Ti -
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OBJECTIVE

The effects of irradiation at low temperatures (nominally 100 to 275°C) on the properties of V-4Cr-4Ti have been examined to define the lower operating temperature limits for this alloy.

SUMMARY

Irradiation at low temperatures (100 to 275°C) to 0.5 dpa causes significant embrittlement and changes in the subsequent room temperature tensile properties of V-4Cr-4Ti. The yield strength and microhardness at room temperature increase with increasing irradiation temperature. The tensile flow properties at room temperature show large increases in strength and a complete loss of work hardening capacity with no uniform ductility. Embrittlement, as measured by an increase in the ductile-to-brittle transition temperature, increases with increasing irradiation temperature, at least up to 275°C. This embrittlement is not due to pickup of O or other interstitial solutes during the irradiation.

INTRODUCTION

Vanadium alloys are candidate materials for structural applications in fusion reactors, due to their low activation, high thermal stress figure of merit (high thermal conductivity, moderate strength, and low coefficient of thermal expansion), and compatibility with liquid lithium. Successful application of these alloys will require understanding of the effects of irradiation on the mechanical properties, such as strength, ductility, and toughness.

The U.S. program has identified V-4Cr-4Ti (wt%) as a leading candidate material. Fast reactor experiments at temperatures in the range 425-600°C have demonstrated promising resistance to radiation damage.[1] The present work examines irradiation behavior in the low temperature regime (100-275°C) to define the lower operating temperature limit for this alloy.

EXPERIMENTAL PROCEDURE

The material used for this experiment was taken from the 500 kg heat produced by Teledyne Wah Chang Albany (TWCA) for the U. S. Fusion Program (heat 832665). This material contains approximately 300 ppm O, 85 ppm N, and 80 ppm C (by weight) [2]. This plate was processed by TWCA and supplied in the form of 6.4-mm thick plate that had been annealed for 2 h at 1050°C, in a vacuum better than 10^{-5} torr. Additional material was supplied as ~40% CW sheet, 1 mm thick.

Subsize Charpy specimens were machined from the annealed plate. These specimens were $3.3 \times 3.3 \times 25.4$ mm with a 30° notch, 0.67 mm deep with a 0.08 mm root radius. The notch was oriented for crack growth perpendicular to the rolling direction (L-T orientation).¹ The specimens were annealed for 2 h at 1000°C in vacuum (better than 10^{-7} torr) after machining. The resultant grain size was 16 μ m. Some specimens were fatigue precracked by cyclic loading in 3-point bending in stroke control, so the load would shed automatically as the crack extended. The final load was approximately 130 N, and the final crack length to specimen width ratio (a/W) was nominally 0.5.

¹ Note that these specimens were incorrectly described as having a notch 0.51 mm deep and being oriented for crack growth parallel to the rolling direction in a previous report "Effect of Cr and Ti Contents on the Recovery, Recrystallization, and Mechanical Properties of Vanadium Alloys" by A. N. Gubbi, A. F. Rowcliffe, D. J. Alexander, M. L. Grossbeck, W. S. Eatherly, and L. T. Gibson, Fusion Materials Semiannual Progress Report for Period Ending December 31, 1995, DOE/ER-0313/19, pp. 37-43, Oak Ridge National Laboratory, Oak Ridge, TN 37831.

Small SS-3 sheet tensiles ($0.76 \times 1.52 \times 7.6$ mm gage section) were machined from the cold-rolled sheet. These specimens were oriented in the longitudinal orientation (parallel to the rolling direction) and were annealed for 2 h at 1000°C in vacuum (better than 10^{-7} torr) after machining. The final grain size of both the Charpy and SS-3 specimens was around $16 \mu\text{m}$.

The irradiation capsules were designed for insertion into the core thimble position in the High Flux Beam Reactor at Brookhaven National Laboratory. The experiment consisted of two separate capsules each containing 5 subcapsules (Fig. 1). The irradiation temperatures were varied from 100 to 275°C by varying the width of the gas gap for each subcapsule. The capsules were purged with ultra high-purity He, evacuated with a turbopump (three cycles), and finally filled to 10^5 Pa. Each subcapsule contained 8 Charpy specimens and 4 tensile specimens held against the holder with a roll pin to provide intimate contact between the holder and the specimens to improve the heat transfer. Vanadium alloy tensile specimens were not included in the 275°C capsule. Each subcapsule had one thermocouple that was attached to a Charpy specimen to monitor temperature throughout the irradiation. One subcapsule in each capsule had 2 thermocouples (cf Fig. 1) for continuous temperature measurement and to detect any asymmetry in heat flow distribution.

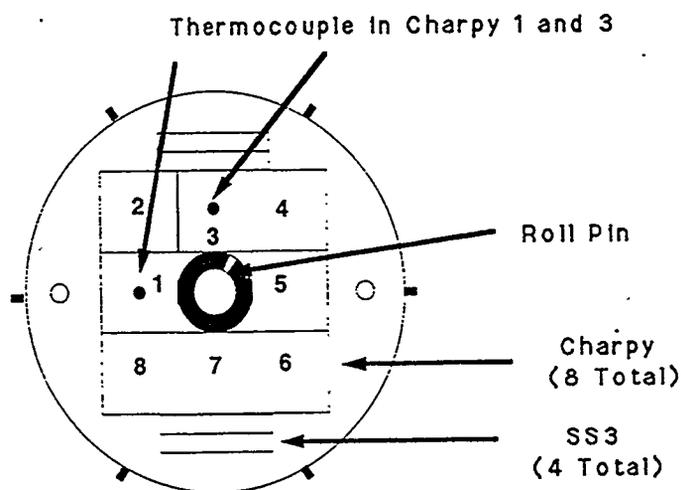


Fig. 1. Schematic illustration of a cross-section of the irradiation capsule, showing location of the subsize Charpy and tensile specimens in a subcapsule.

Each of the two capsules was irradiated for 545 h (23 days) for estimated fast ($E > 0.1$ MeV) and thermal fluences of 8×10^{24} and 3×10^{24} n/m^2 respectively. This dose was calculated to produce about 0.1% Cr and a damage level of 0.4 dpa. Flux monitors in the capsule will be analyzed in the future to provide a more accurate determination of the fluence. It was noticed that the temperature typically increased about 10°C during the course of the irradiation, although the reason for this increase is not clear. The temperatures recorded by the two thermocouples in each subcapsule differed by $<10^\circ\text{C}$ for each irradiation temperature.

The subsize Charpy specimens (both blunt notch and precracked) were tested in air on a pendulum machine modified for small specimens. The tensile specimens were all tested in air at room temperature, with a single test for each irradiation temperature. The tensile tests were conducted on a servohydraulic machine at an initial strain rate of 10^{-3} s^{-1} . The load vs crosshead displacement test record was used to determine the tensile properties.

Vickers hardness measurements were made at room temperature with a 500 g load using the grip-section of SS-3 specimens prior to tensile testing; 2 specimens were tested in each condition, with at least 20 VHN measurements on each specimen.

The room temperature resistivity of the unirradiated control and irradiated tensile specimens was measured prior to tensile testing, using standard 4-point probe techniques (ASTM B 193-87, Standard Test Method for Resistivity of Electrical Conductor Materials, reapproved 1992). An electrical current of 100 mA was supplied by a Keithley model 237 Source Measure Unit through spring-loaded electrical contacts located in the end tab regions of the specimens. The potential drop in the gage region of the specimen was measured between two spring-load electrical contacts that were separated by a distance of 7.1 mm with a Keithley model 182 Sensitive Digital Voltmeter with a low thermal connector (resolution limit of 1 nV). Potentials associated with thermal emfs in the electrical leads were subtracted by using the "relative reading" function of the model 182 voltmeter. Three different tensile specimens were measured for each irradiation temperature. The typical measured resistances were ~1.5-1.8 m Ω . The gage dimensions were measured to an accuracy of ± 2 μ m in two different locations using a Mitutoyo digital micrometer in order to convert the resistance measurement to resistivity values. The experimental error in the resistivity measurements was mainly due to uncertainties in the gage cross-sectional area; the typical measured standard error was ± 0.7 n Ω -m. The temperature was recorded for each measurement (24.5-26°C), and the data were corrected to a reference temperature of 20°C using the V-Ti-Cr alloy resistivity temperature coefficient [3, 4, 5] of 0.75 n Ω -m/K.

RESULTS AND DISCUSSION

The room temperature yield strength and hardness are increased significantly by irradiation from 100 to 275°C (Fig. 2). The increase in yield strength and hardness increases with increasing irradiation temperature, and the yield strength and hardness show similar responses to irradiation. In addition, the tensile flow properties at room temperature change significantly. For each irradiation temperature there is a complete loss of work hardening capability (Fig. 3). Following yielding, the material fails rapidly through plastic instability.

Irradiation at these temperatures (100 to 275°C) produces a very large increase in the ductile-to-brittle transition temperature (DBTT) and a large decrease in the upper-shelf energy (USE) (see Fig. 4). The shift in the DBTT increases with increasing irradiation temperature.

The precracked specimens have higher DBTTs than the blunt notch specimens irradiated at the same temperature. The USE is much lower, primarily due to the greatly reduced cross-sectional area for the precracked specimens.

The existence of a radiation-damage regime associated with rapid hardening and susceptibility to brittle cleavage failure is a common characteristic of BCC metals and alloys. However, the magnitude of the property changes in the V-4Cr-4Ti alloys were larger than those observed, for example, in many reduced activation ferritic-martensitic steels irradiated at ~250°C [6]. Other factors which could contribute to the observed embrittlement are (a) the inadvertent introduction of hydrogen during pre or postirradiation handling and (b) the pick-up of oxygen and nitrogen from the capsule atmosphere during irradiation. These are considered remote possibilities; hydrogen-induced embrittlement has never been observed as a result of the handling procedures used for unirradiated specimens at ORNL and the irradiation temperatures are too low for significant transfer of interstitials into the specimen interior to occur. Nevertheless, two experiments were carried out to test these possibilities.

Several irradiated Charpy specimens were annealed at 400°C for 1 hr in a vacuum of 10^{-7} torr. These conditions were chosen to remove hydrogen while minimizing recovery of radiation damage. The results of subsequent Charpy testing of these specimens are indicated by the solid symbols in Fig. 5. It can be seen that vacuum annealing did not result in significant recovery of impact properties, and it is concluded that hydrogen pick-up is not a significant factor in the observed embrittlement.

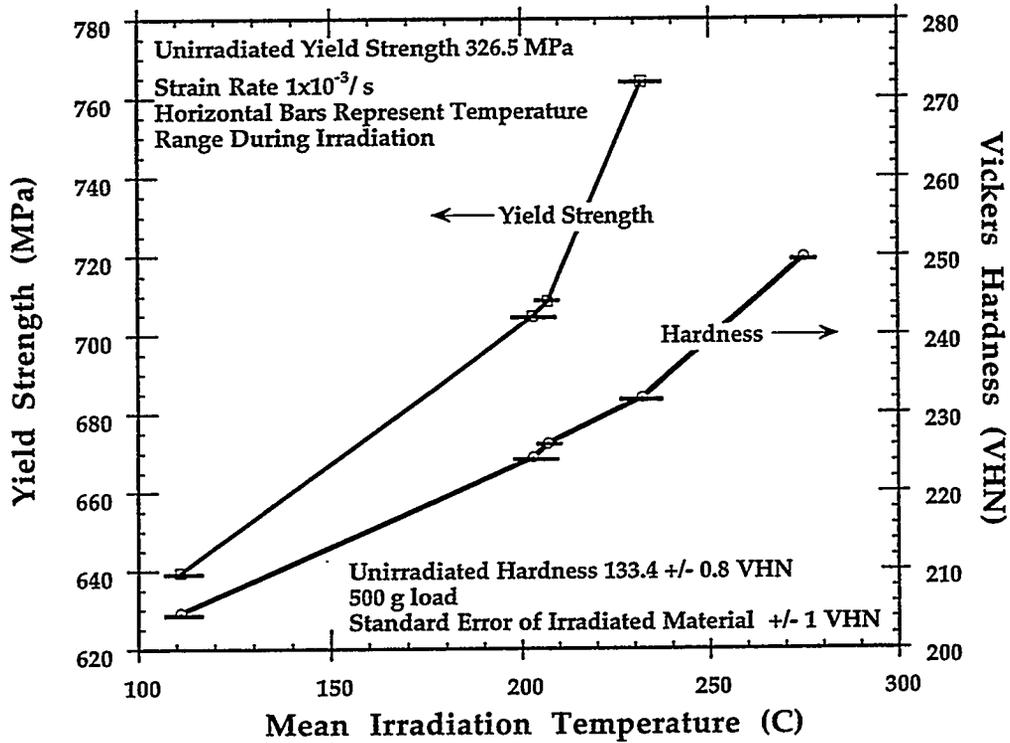


Fig. 2. Effect of irradiation on yield strength and microhardness, measured at room temperature.

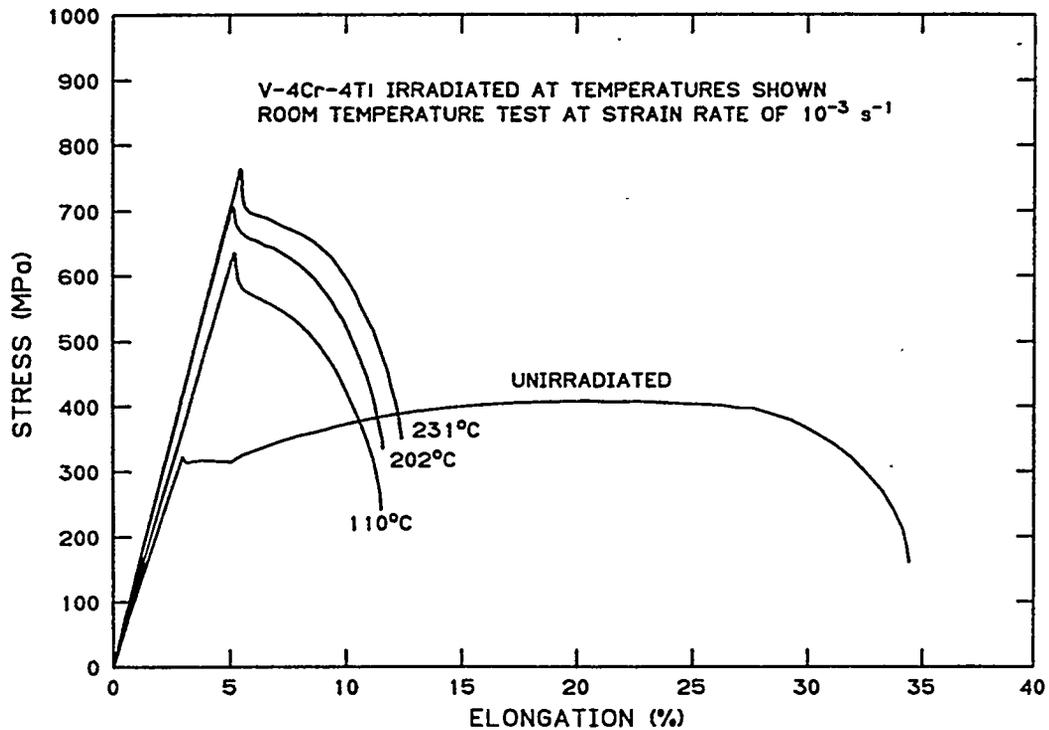


Fig. 3. Stress-elongation curves measured at room temperature, showing the significant decrease in total elongation and the complete loss of work hardening capacity following irradiation.

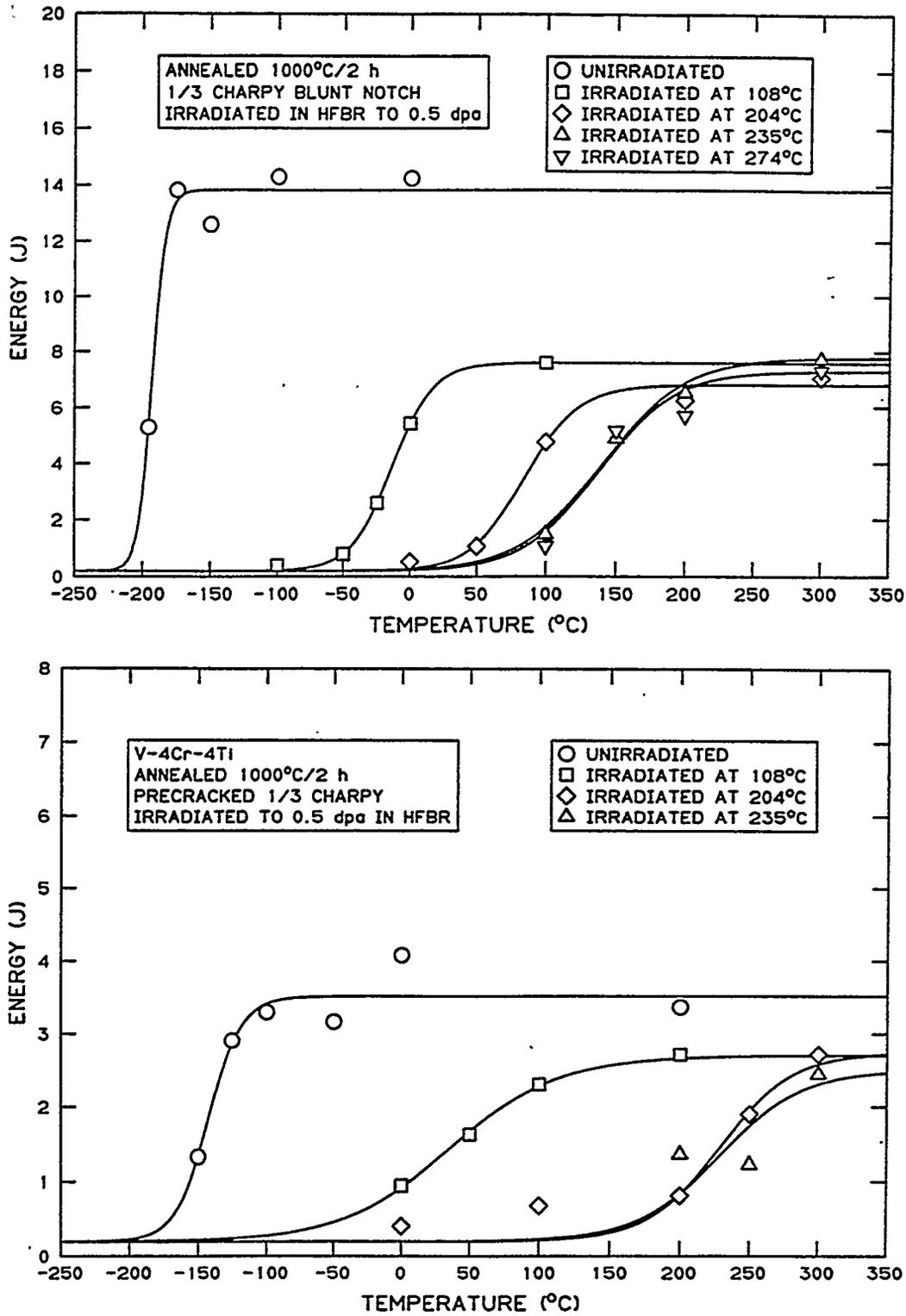


Fig. 4. Impact properties of V-4Cr-4Ti, showing significant embrittlement due to irradiation. Top: blunt notched specimens; bottom: fatigue precracked specimens.

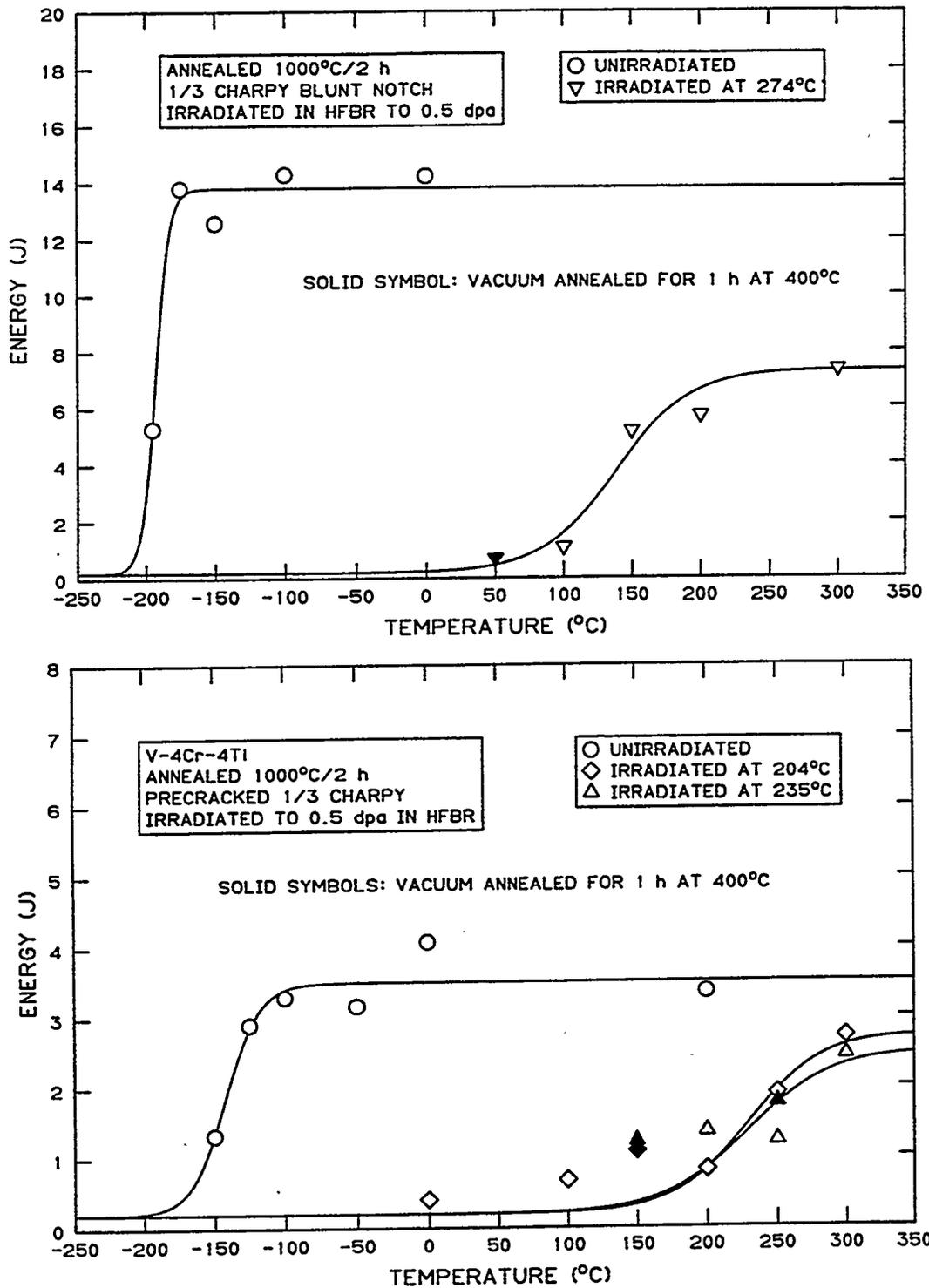


Fig. 5. A comparison of impact properties of specimens after annealing 1 h at 400°C in vacuum with as-irradiated specimens. Annealing had little effect on the impact properties. Top: blunt notched specimens; bottom: precracked specimens.

In a second experiment, one set of unirradiated pre-cracked specimens was placed in aluminum capsules and helium back-filled using procedures identical to those used for the sub-capsules for the irradiation experiment. These capsules were then held at 275°C or 400°C for 21 days. This exposure was sufficient to produce surface oxide films on both sets of specimens. The results of impact tests on these specimens are shown in Fig. 6. At the lower temperatures, impact energies were somewhat above those for the as-annealed specimens, whereas at the higher temperatures, impact energies were slightly lower. However, it is quite clear that the helium-exposed control specimens do not exhibit the completely brittle behavior of the irradiated specimens tested over the same temperature range.

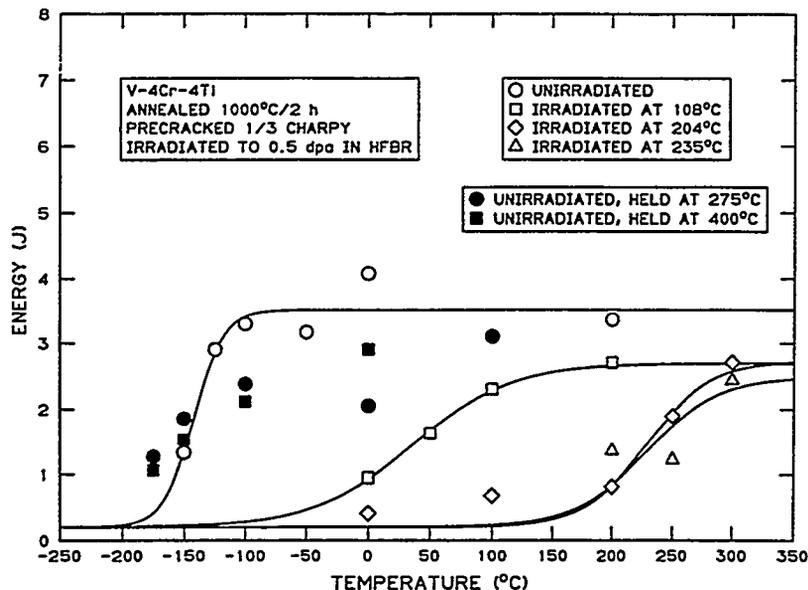


Fig. 6. Impact properties of precracked unirradiated specimens after holding for 21 days at 275 or 400°C in He atmosphere, as compared to unirradiated specimens. Holding at 275 or 400°C had little effect on the impact properties, indicating that no embrittling interstitials were picked up from the He atmosphere.

The room temperature resistivity measurements provide further evidence that interstitial elements are not creating the embrittlement. The room-temperature resistivity increased for irradiation at 108°C as compared to the unirradiated materials (see Fig. 7). This increase in resistivity is solely due to generation of point defect clusters in the lattice, since this temperature is too low for O, C, or N migration in vanadium. The measured migration temperatures and migration enthalpies of C, O, and N in vanadium for 1 h isochronal anneals are ~185°C (1.18 eV), ~185°C (1.26 eV) and ~275°C (1.48 eV) [7]. Similar activation energies have been obtained for oxygen diffusion in V-4Cr-4Ti alloys [8]. After irradiation to 0.4 dpa at ~200°C, the resistivity was similar to the unirradiated value. Electron microscopy studies performed on vanadium irradiated to ~0.1 dpa have found that the defect cluster density decreased by about an order of magnitude as the irradiation temperature increased from 70 to 200°C [9]. Therefore, the lower resistivity of the HFBR specimens irradiated at ~200°C compared to 108°C is likely mainly due to a reduced defect cluster density. In addition, C and O interstitial solutes have moderate mobility at these temperatures [7]. These solutes could migrate to the remaining defect clusters and increase the dislocation barrier strength of the clusters, in a manner analogous to radiation anneal hardening. This proposed interstitial solute strengthening of defect clusters is a possible explanation for the observed increase in microhardness and tensile strength of vanadium irradiated at 200°C compared to 108°C. The observation that the resistivity did not increase dramatically at an irradiation temperature of ~200°C indicates that the observed increase in radiation hardening was not due to solution hardening from pickup of O or C interstitial solutes from the surrounding environment. The specific resistivities for O, C, and N solutes in vanadium are ~54 to 87 nΩ-m/at.% solute [7]. Therefore, the incorporation of significant (>1000 appm) concentrations of O or C in the matrix as solid solution impurities would have caused an easily detected (>5 nΩ-m) increase in the resistivity. The resistivity measured for an irradiation temperature of 240°C was less than that measured in unirradiated

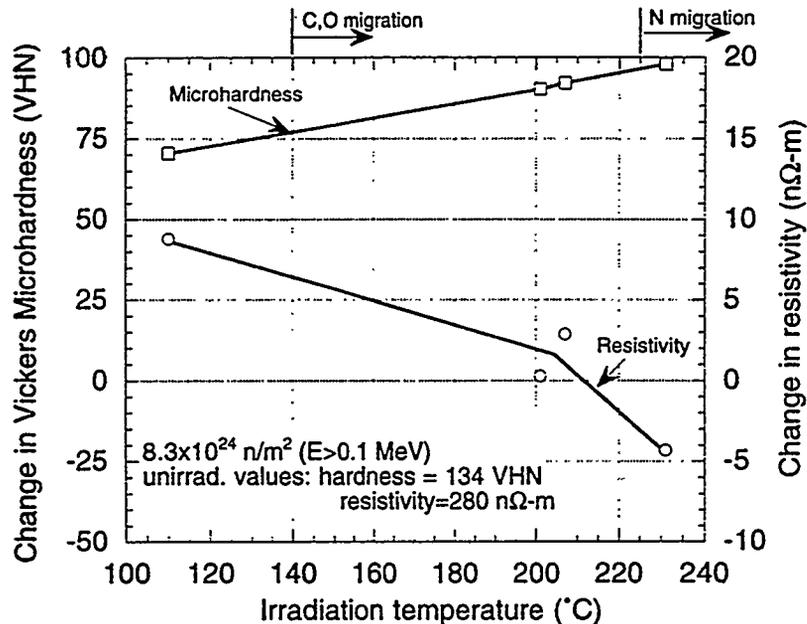


Fig. 7. Resistivity and microhardness after irradiation, as measured at room temperature.

controls. One possible explanation for this result is that ballistic dissolution of the submicroscopic precipitates containing Ti-(O,C,N) has occurred, and that many of the O, C, and N interstitials have migrated to defect clusters. This process would cause a reduction in the measured resistivity compared to the unirradiated alloy if the defect cluster density was much lower than the initial submicroscopic precipitate density, since the resistivity per solute atom should decrease with increasing size. This process would also explain the observed increase in radiation hardening at 240°C compared to the 200°C irradiation (increased barrier strength of defect clusters containing interstitial solute atoms). Additional work, including transmission electron microscopy, is needed to verify this proposed explanation for the temperature-dependent radiation hardening behavior of V-4Cr-4Ti alloys.

SUMMARY AND CONCLUSIONS

It is clear from this experiment that irradiation to 0.5 dpa at low temperatures (100 to 275°C) causes significant embrittlement of V-4Cr-4Ti. This embrittlement is not due to pickup of interstitial solutes during the irradiation, but is directly related to radiation hardening. The degree of embrittlement increases with increasing irradiation temperature, at least up to 275°C. The resistivity results suggest that the effectiveness of defect clusters as barriers to dislocation motion is enhanced by the migration of oxygen and carbon to defect clusters at temperatures above ~200°C.

Irradiation at these low temperatures causes significant changes in the subsequent tensile flow properties at room temperature, with large increases in strength and a complete loss of work hardening capacity with no uniform ductility. The loss of work hardening may be related to dislocation channeling; further work is needed to characterize the defect microstructure produced during irradiation to identify the mechanism of hardening. The range of temperatures over which embrittlement occurs needs to be determined, and further irradiation experiments at 330 and 400°C are in preparation.

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