EFFECT OF DYNAMICALLY CHARGED HELIUM ON TENSILE PROPERTIES OF V–5Ti, V–4Cr–4Ti, AND V–3Ti–1Si* H. M. Chung, B. A. Loomis, L. Nowicki, and D. L. Smith (Argonne National Laboratory)

SUMMARY

In the Dynamic Helium Charging Experiment (DHCE), helium was produced uniformly in the specimen at linear rates of ≈0.4 to 4.2 appm He/dpa by the decay of tritium during irradiation to 18–31 dpa at 425–600°C in the lithium-filled DHCE capsules in the Fast Flux Test Facility. This report presents results of postirradiation tests of tensile properties of V–5Ti, V-4Cr-4Ti, V–3Ti–1Si. The effect of helium on tensile strength and ductility was insignificant after irradiation and testing at >420°C. Contrary to initial expectation, room–temperature ductility of DHCE specimens was higher than that of non–DHCE specimens, whereas strength was lower, indicating that different types of hardening centers are produced during DHCE and non–DHCE irradiation. In strong contrast to results of tritium–trick experiments, in which dense coalescence of helium bubbles is produced on grain boundaries in the absence of displacement damage, no intergranular fracture was observed in any tensile specimens irradiated in the DHCE.

INTRODUCTION

Recent attention in the development of vanadium alloys has focused on V-4Cr-4Ti for fusion reactor structural components because of its excellent mechanical and physical properties before and after neutron irradiation.[1,2] Similar to V–4Cr–4Ti, the V–5Ti and V–3Ti–1Si also exhibited excellent resistance to irradiation embrittlement under non–DHCE conditions (negligible helium generation).[2] However, one property of these alloys that is not well understood is the effect of helium; only initial results of tensile testing on V–4Cr–4Ti have been reported on the effects of simultaneous generation of helium and neutron displacement damage under fusion–relevant conditions (i.e., ≈5 appm He/dpa ratio), although the effect of helium on other vanadium alloys has been investigated by less–than–prototypical simulation techniques such as tritium-trick,[3,4] cyclotron-injection,[5] and boron-doping.[5,6] In the DHCE,[7,8] fusion–relevant helium–to–dpa damage ratio is closely simulated by utilizing slow transmutation of controlled amounts of ^{6}Li and a tritium–doped mother alloy immersed in ^{6}Li + ^{7}Li. This report presents results of postirradiation examination of mechanical properties of V–5Ti, V-4Cr-4Ti, and V–3Ti–1Si alloys, which have been reported to exhibit excellent resistance to displacement damage under non–DHCE irradiation.[2]

MATERIALS AND PROCEDURES

The elemental composition of the V–5Ti, V–4Cr–4Ti, and V–3Ti–1Si alloys, determined prior to irradiation, is given in Table 1. The alloy ingots, melted from low–chlorine titanium and high–purity vanadium by the multiple–vacuum–arc process, were typically extruded at 1150°C and annealed at 1050°C. Final forms of the product were 3.8–, 1.0–, and 0.3–mm–thick annealed plates and sheets. Tensile specimens with a gauge length of 7.62 mm and a gauge width of 1.52 mm were machined from 1.0–mm–thick sheets that had been annealed at ≈1050°C. The only secondary phase in the recrystallized specimen was Ti(O,N,C), which is normally observed in titanium–containing vanadium alloys with >400 wppm O+N+C. Tensile properties were measured at 23°C and at irradiation temperatures in flowing argon at a strain rate of 0.0011 s^{-1}. The thickness and gauge width of each specimen were measured individually after irradiation and before each tensile test.

The alloy specimens were irradiated in the Fast Flux Test Facility (FFTF) at 420, 520, and 600°C to neutron fluences (E > 0.1 MeV) ranging from 3.7 x 10^{22} n/cm^2 (≈18 displacements per atom, or dpa) to 6.4 x 10^{22} n/cm^2 (≈31 dpa). Helium in the alloy specimens was produced by transmutation of controlled amounts of ^{6}Li and predetermined amounts of tritium–doped vanadium mother alloy immersed in ^{6}Li + ^{7}Li.[7,8] Table 2 summarizes the irradiation temperature, weight of the mother alloy, fraction of ^{6}Li, and tritium and lithium inventory charged in each of the seven DHCE capsules before irradiation.

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Table 1. Composition of vanadium–base alloys

ANL ID	Nominal Composition (wt.%)	Impurity Composition (wppm) O	N	C	Si
BL-45	V–2.5Ti–1Si	345	125	90	9900
BL-46	V–4.6Ti	305	53	85	160
BL-47	V–4.1Cr–4.3Ti	350	220	200	870

Table 3 summarizes actual postirradiation parameters determined from tensile and transmission electron microscopy disk specimens of the V–4Cr–4Ti alloy, i.e., fast neutron fluence, dose, and helium and tritium contents measured ≈20–25 days after the postirradiation tests. Helium and tritium content was determined by mass spectrometry at Rockwell International Inc., Canoga Park, CA. The tritium content was determined on the basis of an analysis of ^3He decay, measured on the same specimens ≈50 days apart.

Table 2. Summary of capsule–loading parameters of DHCE

Capsule ID No.	Irradiation Temp. (°C)	Total Weight (g) Vanadium [a]	Specimen [b]	Lithium	Fraction of ^6Li (%)	Initial Tritium Charged [a] (Ci)	(mmol)
4D1	425	1.5468	5.86	0.765	5.0	99	1.70
4D2	425	1.5536	5.38	0.765	4.5	70	1.20
5E2	425	1.5657	5.38	0.670	1.0	26	0.45
5D1	500	1.5727	5.77	0.938	6.5	73.5	1.26
5E1	500	1.5651	5.82	0.952	1.0	57	0.98
5C1	600	1.5656	5.82	0.808	8.0	16.4	0.28
5C2	600	1.5466	5.95	0.955	8.0	18	0.31

[a] Letter from C. E. Johnson to K. Pearce, April 23, 1991; 1 mmol = 58.3 Ci.
[b] Excluding tritium–charged mother alloy.

Table 3. Summary of irradiation parameters of Dynamic Helium Charging Experiment and helium and tritium contents measured in V–4Cr–4Ti specimens

Capsule ID No.	Irradiation Temp. (°C)	Fluence (E >0.1 MeV) (10^{22} n cm^{-2}	•Total Damage (dpa)	Calculated Helium (appm) to dpa Ratio[a] at EOI[b] (Assumed k_a or k_w)[c] k_a=0.073 (k_w=0.01)	Measured Helium Content[d] (appm)	Actual Helium to dpa Ratio (appm/dpa)	Measured Tritium Content[e] (appm)
4D1	425	6.4	31	3.8	11.2–13.3	0.39	27
4D2	425	6.4	31	2.8	22.4–22.7	0.73	39
5E2	425	3.7	18	2.1	3.3–3.7	0.11	2
5D1	500	3.7	18	4.4	14.8–15.0	0.83	4.5
5E1	500	3.7	18	3.1	6.4–6.5	0.36	1.7
5C1	600	3.7	18	1.1	8.4–11.0	0.54	20
5C2	600	3.7	18	1.1	74.9–75.3	4.17	63

[a] L. R. Greenwood "Revised Calculations for the DHCE," April 30, 1993.
[b] Beginning of irradiation (BOI) May 27, 1991; end of irradiation (EOI) March 19, 1992; 203.3 effective full power days (EFPD), hot standby at ≈220°C until November 1992.
[c] Equilibrium ratio (k_a by atom, k_w by weight) of tritium in V alloy to that in surrounding liquid lithium.
[d] Measured June 1994.
[e] Measured August 1994.

RESULTS

Yield strength, ultimate tensile strength, uniform elongation, and total elongation, measured on tensile specimens of V–4Cr–4Ti irradiated at 425°C–600°C to 18–34 dpa in the DHCE (4–75 appm He, or helium generation rate of ≈0.4–4.2 appm He/dpa), are summarized in Fig. 1. For comparison, similar properties, measured on irradiated non–DHCE (<0.1 appm He) specimens, are also plotted as a function of irradiation temperature. Similar results from V–5Ti (9–20 appm He) and V–3Ti–1Si (6–36 appm He) are given in Figs. 2 and 3, respectively. In these figures, two groups of postirradiation tensile properties are given, i.e., those measured at the irradiation temperatures and those measured at 20–200°C.

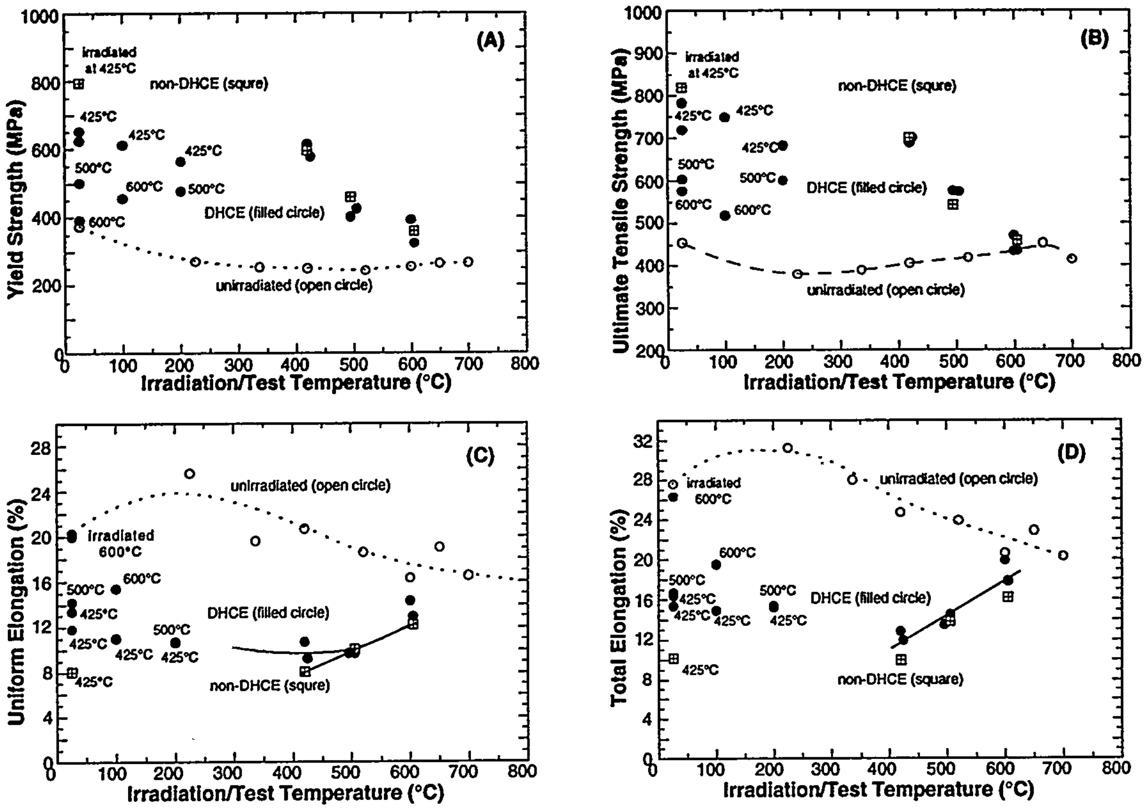

Fig. 1. Yield strength (A), ultimate tensile strength (B), uniform elongation (C), and total elongation (D) of V–4Cr–4Ti after irradiation at 420–600°C to 18–34 dpa in DHCE (0.4–4.2 appm He/dpa) and non–DHCE conventional irradiation (negligible helium generation).

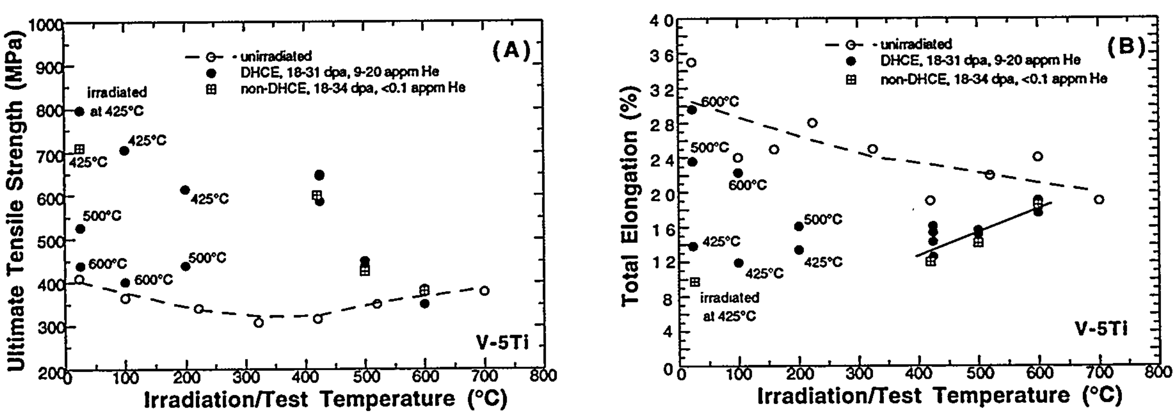

Fig. 2. Ultimate tensile strength (A) and total elongation (B) of V–5Ti after irradiation at 420–600°C to 18–34 dpa in DHCE (9–20 appm He) and non–DHCE conventional irradiation (<0.1 appm He).

After irradiation to ≈30 dpa in either a DHCE or a non–DHCE, ductilities of all three alloys remained significantly high, i.e., >9% uniform elongation and >12% total elongation. Tensile properties of DHCE specimens, measured at 425°C, 500°C, and 600°C (the same as the irradiation temperatures), were essentially the same as or slightly higher than those measured on non–DHCE specimens, showing that the effect of helium was insignificant. For all three alloys, ductility at 20–200°C of the DHCE specimens (irradiated at 425°C, 500°C, and 600°C) was higher than that of similar non–DHCE specimens, whereas strength was lower. This was an unexpected finding. Initially, this finding was reported for V–4Cr–4Ti; the present confirmation of similar behavior in V–5Ti and V–3Ti–1Si provides a conclusive trend. Although the mechanisms leading to the higher ductility and lower strength of the DHCE specimens are not understood at this time, the consistent observations indicate that different types of hardening centers are produced during DHCEs and non–DHCEs.

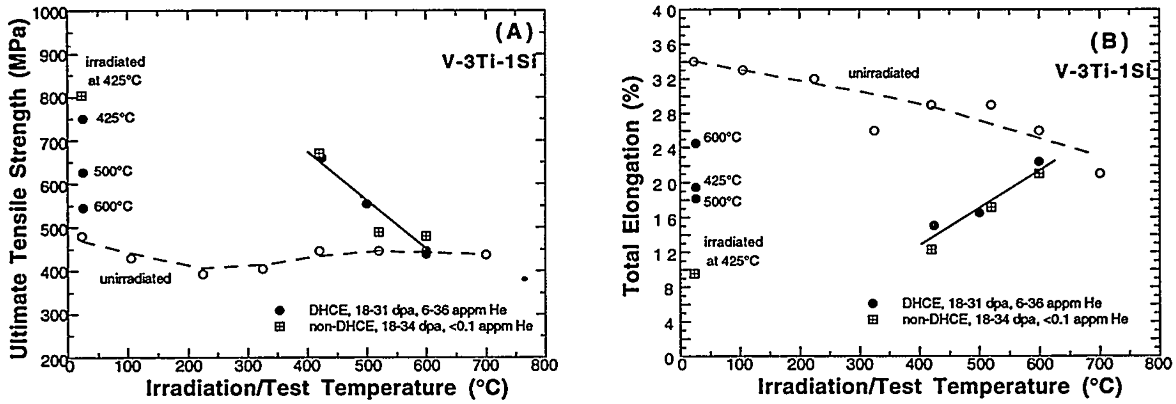

Fig. 3. Ultimate tensile strength (A) and total elongation (B) of V–3Ti–1Si after irradiation at 420–600°C to 18–34 dpa in DHCE (6–36 appm He) and non–DHCE conventional irradiation (<0.1 appm He).

The dependence of uniform and total elongation on irradiation and test temperature, is in sharp contrast to similar results obtained from specimens in which helium atoms were produced by the tritium–trick method. In the latter type experiments, total elongation measured at room temperature and at 700–800°C was significantly lower than that measured at 500–600°C, because of the strong susceptibility to intergranular cracking that is associated with extensive formation of grain–boundary helium bubbles.[4] However, no intergranular fracture surface morphology was observed in the tensile specimens of V–5Ti, V–4Cr–4Ti, and V–3Ti–1Si irradiated in the DHCE and tested at 25–600°C (including the V–4Cr–4Ti specimen irradiated in Capsule 5C2 at 600°C at a helium generation rate of 4.2 appm He/dpa), and no ductility degradation similar to that in tritium–trick experiments was observed. This is shown in Fig. 4, where the ratio of total strain in specimens with and without helium is plotted as a function of irradiation and test temperature for the DHCE and tritium–trick experiments.

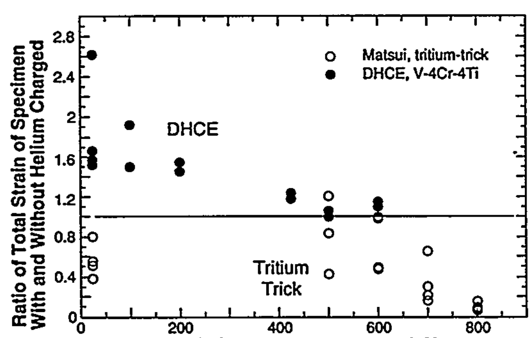

Fig. 4.
Ratio of total strain in specimens with and without helium as a function of irradiation and test temperature. Results obtained from tritium–trick experiment and DHCE are shown for comparison.

DISCUSSION

An important finding from the DHCE was that the actual (measured) amount of helium and tritium in V–5Ti, V–4Cr–4Ti, and V–3Ti–1Si specimens was significantly lower than previously calculated (see Table 3) on the basis of an assumed equilibrium ratio ($k_W = 0.01$, by weight) of tritium in the alloys to that in the liquid lithium (Table 2). In the case of V–4Cr–4Ti, except for specimens irradiated in Capsule 5C1 and 5C2 at 600°C, actual helium/dpa ratios (i.e., 0.36–0.83) were several times lower than those calculated on the basis of an equilibrium ratio of $k_W = 0.01$ (i.e., 2.1–4.4). The helium generation rates in the specimens in Capsule 5C2 (≈4.2 appm He/dpa) were, however, close to the fusion-relevant rate of ≈5 appm He/dpa. The smaller helium/dpa rate, in particular for irradiation at ≈420°C, indicates that the level of hydrogen and tritium in the lithium-cooled V–4Cr–4Ti first-wall/blanket structure, and hence the effect of hydrogen and tritium on fracture toughness, will be significantly lower than previously assumed.

Helium microvoids were insignificant in all of the specimens that were irradiated at 500–600°C in the DHCE; only a few helium bubbles were observed at the interface between the grain matrix and some Ti(O,N,C) precipitates that are normally present in Ti-containing alloys. Even in V–4Cr–4Ti specimens that were irradiated at 600°C at the highest helium generation rate of ≈4.2 appm helium/dpa (Capsule 5C2), no microvoids could be detected in either grain matrix or grain boundaries.[9] For V–4Cr–4Ti specimens irradiated to 31 dpa at 425°C, low densities of diffuse helium bubbles were distributed more or less uniformly in a localized grain matrix and near a limited fraction of grain boundaries.

Although, in some of the V–5Ti and V–3Ti–1Si specimens irradiated at 425°C, more helium bubbles were observed in limited regions of grain boundaries than in grains, significant coalescence of helium bubbles, which is nearly continuous on grain boundaries in tritium-tricked specimens, was not observed. However, considering the relatively low helium generation rates measured in the specimens irradiated at ≈425°C, it is desirable to demonstrate the above finding for higher helium generation rates (e.g., 5–7 appm He/dpa) from a modified DHCE at 300–450°C. Neither partially nor fully intergranular failure was observed in any of the present specimens that were irradiated in the DHCE and subsequently tested in uniaxial tension at 20–600°C. These observations, consistent with the characteristics of the helium bubble distribution described above, are also in distinct contrast to the propensity for intergranular failure that is commonly observed in tritium-trick experiments.

For the three alloys, V–5Ti, V–4Cr–4Ti, and V–3Ti–1Si, the uniform and total elongations, determined from the tensile tests at 20–200°C on DHCE specimens, were significantly greater than similar elongations measured on specimens irradiated in either non-DHCE (Figs. 1–3) or tritium-trick experiments (Fig. 4). This is also consistent with the absence of continuous aggregation of helium bubbles on grain boundaries in the specimens irradiated in the DHCE. In addition, the observation indicates that different types of hardening centers are produced in the alloy during DHCE and non-DHCE irradiation at 425–600°C.

Although the nature of the hardening centers produced during DHCE is not understood at this time, we believe that helium atoms are involved. In a series of studies on thermal desorption of helium from unalloyed vanadium and V–5Ti irradiated with helium ions of various energy levels, van Veen et al.[10] and Buitenhuis et al.[11] concluded that helium-oxygen-vacancy and helium-nitrogen-vacancy (and probably helium-vacancy-carbon as well) complexes are formed in the irradiated material. These investigators further deduced that the complexes are stable at low temperatures (<230°C) but dissociate into helium atoms and oxygen-vacancy and nitrogen-vacancy complexes at 270–310°C, leading to a prominent helium desorption peak at ≈290°C that was observed consistently in their experiments. Desorption peaks at ≈770°C and ≈1250°C, observed only after irradiation with helium ion to higher doses, were attributed to clusters of helium atoms and helium bubbles, respectively. These clusters and bubbles of helium are believed to be unstable only at the high temperatures. During the degassing treatment in the present study in which DHCE specimens were heated to 400°C at a rate of ≈0.2°C/s, desorption peaks were observed consistently at ≈290°C, although helium desorption was not positively identified by mass spectroscopy, as done by van Veen et al.[10] and Buitenhuis et al.[11]

Based on these observations, it is likely that stable helium-vacancy-impurity complexes are also present in the specimens irradiated in DHCE during tensile tests at room temperature. In contrast, in specimens irradiated in non-DHCE under similar conditions, vacancies and impurities (such as oxygen, nitrogen, and carbon) are not expected to form complexes in the absence of appreciable helium atoms. Rather, the impurity atoms in solution and vacancies or vacancy clusters will be scattered more or less randomly in interstitial and vacancy sites, respectively. Dislocation motion would then be more difficult, and hence ductility would be lower in the non-DHCE than in the DHCE specimens; this seems to be in accordance with the results shown in Figs. 1–3.

CONCLUSIONS

1. Tensile ductilities of V–5Ti, V–4Cr–4Ti, and V–3Ti–1Si, irradiated to 18–31 dpa at 425°C to 600°C in the Dynamic Helium Charging Experiment (DHCE) at helium generation rates of 0.4–4.2 appm He/dpa, remained significantly high at 25–600°C, i.e., >9% uniform elongation and >12% total elongation. Tensile properties measured at >400°C were essentially the same as those measured on non–DHCE specimens (negligible helium), showing that the effects of helium were insignificant.

2. Postirradiation ductility of these alloys, measured at 20–200°C, was higher than that of similar non–DHCE specimens, whereas strength was lower. These observations indicate that different types of hardening centers are present at room temperature in the DHCE specimens (helium–vacancy–impurities complex, impurities being oxygen, nitrogen, and carbon) and in non–DHCE specimens (defects and defect clusters, impurities in interstitial sites).

3. Fracture morphology and the dependence of uniform and total elongation on irradiation and test temperature were in distinct contrast to similar results obtained from specimens in which helium atoms were produced by the tritium–trick method. Neither partial nor predominantly intergranular fracture was observed in tensile specimens irradiated in the DHCE and tested at 20–600°C.

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