EFFECT OF FAST NEUTRON IRRADIATION TO 4 DPA AT 400°C ON THE PROPERTIES OF V-(4-5)Cr-(4-5)Ti ALLOYS — S. J. Zinkle, D. J. Alexander, J. P. Robertson, L. L. Snead, A. F. Rowcliffe, L. T. Gibson, W. S. Eatherly (Oak Ridge National Laboratory) and H. Tsai (Argonne National Laboratory)

OBJECTIVE

The objective of this report is to summarize tensile, Charpy impact and electrical resistivity measurements on V-4Cr-4Ti and V-5Cr-5Ti alloys that were irradiated in the EBR-II reactor at ~400°C to a dose of about 4 dpa.

SUMMARY

Tensile, Charpy impact and electrical resistivity measurements have been performed at ORNL on V-4Cr-4Ti and V-5Cr-5Ti specimens that were prepared at ANL and irradiated in the lithium-bonded X530 experiment in the EBR-II fast reactor. All of the specimens were irradiated to a damage level of about 4 dpa at a temperature of ~400°C. A significant amount of radiation hardening was evident in both the tensile and Charpy impact tests. The irradiated V-4Cr-4Ti yield strength measured at ~390°C was >800 MPa, which is more than three times as high as the unirradiated value. The uniform elongations of the irradiated tensile specimens were typically ~1%, with corresponding total elongations of 4-6%. The ductile to brittle transition temperature of the irradiated specimens as determined by dynamic Charpy impact tests was ≥300°C. The electrical resistivity of the irradiated specimens was less than the unirradiated resistivity, which suggests that hardening associated with interstitial solute pickup was minimal.

PROGRESS AND STATUS

Introduction

Vanadium alloys with solute contents of 4-5%Cr and 4-5%Ti have recently been identified as promising candidates for the first wall structure of magnetic fusion energy systems [1,2]. A 500 kg production heat of V-4Cr-4Ti (heat #832665) was recently fabricated in order to perform a comprehensive evaluation of the potential and limitations of this alloy composition. Very favorable mechanical properties were measured for the unirradiated alloy, including ductile to brittle transition temperatures (DBTTs) of about -150°C and -200°C as determined from Charpy vee-notch (CVN) impact tests on precracked and machined notch specimens, respectively [3].

Several fast reactor irradiation experiments have reported that the vanadium alloys containing 4-5% Cr and 4-5% Ti exhibit exceptional resistance to radiation embrittlement at doses of ~18 to 30 dpa at temperatures between 425 and 600°C [1,4-7]. Although radiation hardening increased the yield strength to as high as ~600 MPa at 430°C, the uniform elongations were reported to be >8% for all irradiation conditions and the Charpy impact DBTT in irradiated specimens was reported to remain near -200°C. It was recently discovered that the tensile elongations for these experiments were incorrectly analyzed, and the revised elongations are significantly smaller than previously reported [8]. However, vanadium alloys containing 4-5% Cr and 4-5% Ti still appear to be promising materials for fusion energy structural applications.

A recent low-dose, low-temperature neutron irradiation experiment found that significant radiation hardening and embrittlement occurred in the 500 kg heat of V-4Cr-4Ti following irradiation to a damage level of ~0.4 dpa at 110-275°C [3]. Typical irradiated yield strengths were ~650 MPa, with corresponding uniform elongations of <0.2%. The irradiation caused a dramatic increase in the DBTT, with DBTT values of ~140°C and ~250°C, respectively for machined notch and precracked Charpy specimens irradiated at 235°C. Another study has reported significant radiation hardening and reductions in uniform elongation to 2.0% and 3.5% in a V-5%Cr-5%Ti alloy following FFTF irradiation to 49 dpa at 407°C (non-DHCE) and 24 dpa at 430°C (DHCE), respectively [9]. These two studies [3,9] and the revised tensile data set from other reactor irradiation experiments [8] suggest that radiation hardening and concomitant reduction in ductility may be pronounced in V-4Cr-4Ti alloys at irradiation temperatures below 430°C.

The X530 experiment in the EBR-II fast reactor was designed to obtain initial irradiation data on the 500 kg heat of V-4Cr-4Ti at a relatively low temperature of ~400°C. Several different preirradiation heat treatments were included in the specimen matrix in order to investigate effects of thermomechanical treatment on the mechanical properties. Specimens from the BL-63 heat of V-5Cr-5Ti with different thermomechanical treatments were also irradiated. Previous work has shown that the BL-63 heat is susceptible to embrittlement following annealing at temperatures above ~1100°C, whereas good mechanical properties were obtained in unirradiated specimens annealed at temperatures ≤1050°C [10].

Experimental procedure

The present study represents a subset of the specimen matrix that was irradiated in the X530 experiment [11,12]. Miniature machined-notch Charpy (MCVN) impact bar specimens (nominal dimensions 3.3 × 3.3 × 25.4 mm) and type SS-3 miniature sheet tensile specimens (nominal gage dimensions 0.76 × 1.52 × 7.6 mm) were fabricated from the V-4Cr-4Ti and V-5Cr-5Ti alloys. As summarized in Table 1, three different preirradiation heat treatments were investigated for each of the two alloys in the present study.

Table 1. Summary of X530 specimen matrix investigated at ORNL in this study

Alloy	Heat treatment	Specimen types
V-4Cr-4Ti (#832665)	TWCA&950 (Wah Chang anneal/1050°C & annealed 950°C/1h)	MCVN*
"	WR/950 (warm rolled at 400°C and annealed at 950°C/1h)	SS3
"	WR/1050 (warm rolled at 400°C and annealed at 1050°C/1h)	SS3
"	WR/1125 (warm rolled at 400°C and annealed at 1125°C/1h)	SS3
V-5Cr-5Ti (#BL-63)	TWCA ann. (Wah Chang mill anneal at 1050°C)	SS3, MCVN
"	CR/950 (cold rolled at 20°C and annealed at 950°C/1h)	SS3, MCVN
"	CR/1050 (cold rolled at 20°C and annealed at 1050°C/1h)	SS3, MCVN

*previously incorrectly reported as warm rolled at 400°C and annealed at 950°C/1h [11,12]

The specimens were irradiated in Li-bonded type 316 stainless steel subcapsules that were contained in two adjacent flow-through (weeper) capsule tubes in the EBR-II core position 2F1. The calculated specimen temperatures ranged from ~371°C for the subcapsules at the bottom on the core to ~410°C for subcapsules near the top of the core [11]. The tensile specimens for the V-4Cr-4Ti (heat 832665) and V-5Cr-5Ti alloys (BL-63) were located slightly above the horizontal midplane of the core in subcapsule positions S9 and S8, with calculated specimen temperatures of ~400°C and ~405°C, respectively [11,12]. The estimated fast neutron (E>0.1 MeV) fluences for these specimens are ~7.3×10^{25} n/m^2 and ~6.6×10^{25} n/m^2, respectively (~4 and ~3.6 dpa). The Charpy impact specimens for these alloys were distributed in 4 different subcapsules (S7,S3,S6,S5) below and above the horizontal midplane of the core, with calculated specimen temperatures of ~380-410°C and damage levels of ~3.5 to 4 dpa. Further irradiation details are given elsewhere [11,12].

Following irradiation, the specimens were removed from the Li-filled subcapsules at ANL-East and shipped to ORNL for testing. The gage dimensions of the irradiated SS-3 tensile specimens were measured prior to testing. Electrical resistivity measurements were performed at room temperature on the tensile specimens using techniques that are summarized elsewhere [3]. For the elevated temperature tensile tests, the specimens were heated in a vacuum furnace to a temperature of ~390°C (typical ramp time of ~15 min.) and held at the test temperature for 30 minutes prior to the start of the tensile test. The pressure during the tensile testing was 1 to 2.5×10^{-6} torr. One of the WR/950 V-4Cr-4Ti specimens was also tested in laboratory air at room temperature. All of the tensile specimens were tested in a servohydraulic machine at a constant crosshead speed of 0.0085 mm/s, which corresponds to an initial strain rate of 1.1×10^{-3} s^{-1}. The data were recorded digitally and also on a chart recorder. The engineering tensile properties were obtained from analysis of the load vs. crosshead displacement test record. Charpy impact testing was performed in air on a pendulum machine modified for small specimens [3].

Results and discussion

The results of the electrical resistivity measurements are shown in Table 2. The typical experimental standard deviation for the resistivity measurements was ± 1 nΩ-m. There was no significant difference in the measured resistivities for the 3 preirradiation heat treatments of the irradiated V-4Cr-4Ti alloy. However, the resistivity measured in the V-5Cr-5Ti alloy was higher for the cold-rolled and annealed specimens compared to the vendor-annealed specimens. Electrical resistivity measurements have not yet been performed on control specimens from this experiment, so the quantitative change in electrical resistivity associated with the neutron irradiation cannot be determined. Comparison with electrical resistivity measurements [13] obtained on specimens from the 500 kg V-4Cr-4Ti heat with nominally identical heat treatments suggests that the resistivity of the neutron-irradiated specimens was ~10 nΩ-m lower than unirradiated specimens. A large increase in the electrical resistivity of ~5 nΩ-m would have occurred if significant amounts (>0.03 at.%) of interstitial solute (H,O,C,N) were picked up and dissolved in the specimen matrix during or following the irradiation.

Table 3 summarizes the results of the tensile testing. The yield strengths of all of the irradiated specimens tested at the irradiation temperature were between 740 and 880 MPa, which is about three times the unirradiated yield strength at this temperature. The high amount of radiation hardening was accompanied by a dramatic reduction in the uniform elongation, with typical values near 1%. Typical unirradiated uniform elongations for these alloys at 400°C are >15%. Figure 1 shows a representative load vs. crosshead displacement curve for irradiated V-4%Cr-4%Ti that was tested at 390°C. Hardening associated with pickup of interstitial solutes (H,O,C,N) does not appear to be a likely explanation for the reduction in ductility, since the electrical resistivity of the irradiated specimens was apparently less than the unirradiated resistivity. The V-4Cr-4Ti specimens that were annealed at 1125°C prior to irradiation exhibited lower ductility and higher strength compared to specimens annealed at 950 or 1050°C. There was no significant difference between the three preirradiation heat treatment conditions for the V-5Cr-5Ti alloy. The tensile elongations and strengths were all higher for a V-4Cr-4Ti specimen tested at room temperature compared to the 390°C test temperature (Table 3). This type of behavior is commonly observed in metals that have been irradiated in the radiation hardening regime ($T_{irr}<0.3\ T_M$).

Table 2. Summary of electrical resistivity measurements on the irradiated specimens

Alloy	Preirradiation heat treatment	Electrical resistivity at 20°C
V-4Cr-4Ti (#832665)	WR/950	276.2 nΩ-m
"	WR/1050	274.2 nΩ-m
"	WR/1125	274.2 nΩ-m
V-5Cr-5Ti (#BL-63)	TWCA ann.	285.2 nΩ-m
"	CR/950	291.5 nΩ-m
"	CR/1050	293.0 nΩ-m

Table 3. Summary of tensile measurements on the irradiated specimens

Alloy	Preirr. heat treatment	T_{irr}	T_{test}	0.2% yield strength (MPa)	Ultimate strength (MPa)	uniform elongation	Total elongation
V-4Cr-4Ti	WR/950	400°C	390°C	819±21*	847±20*	0.8±0.04%*	4.1±0.2%*
"	WR/1050	400°C	390°C	809	818	0.4%	6.0%
"	WR/1125	400°C	390°C	880	883	0.2%	6.0%
V-5Cr-5Ti	TWCA ann.	405°C	390°C	733	774	1.2%	7.3%
"	CR/950	405°C	390°C	701	743	0.8%	2.8%
"	CR/1050	405°C	390°C	723	765	1.0%	7.0%
V-4Cr-4Ti	WR/950	400°C	20°C	880	935	2.2%	8.5%

*average of 2 specimens

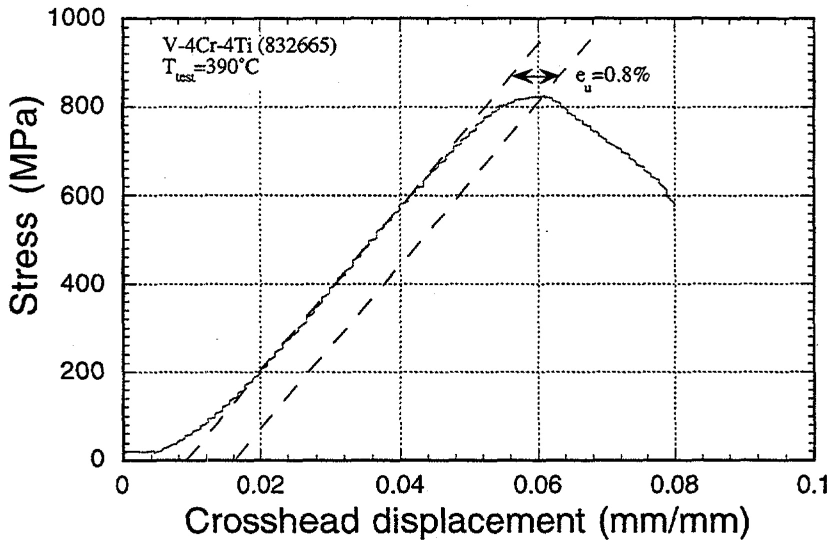

Fig. 1. Load vs. crosshead displacement curve for a warm-worked and 950°C annealed V-4Cr-4Ti specimen (ID# B71/36) that was irradiated to ~4 dpa at ~400°C and tested at 390°C.

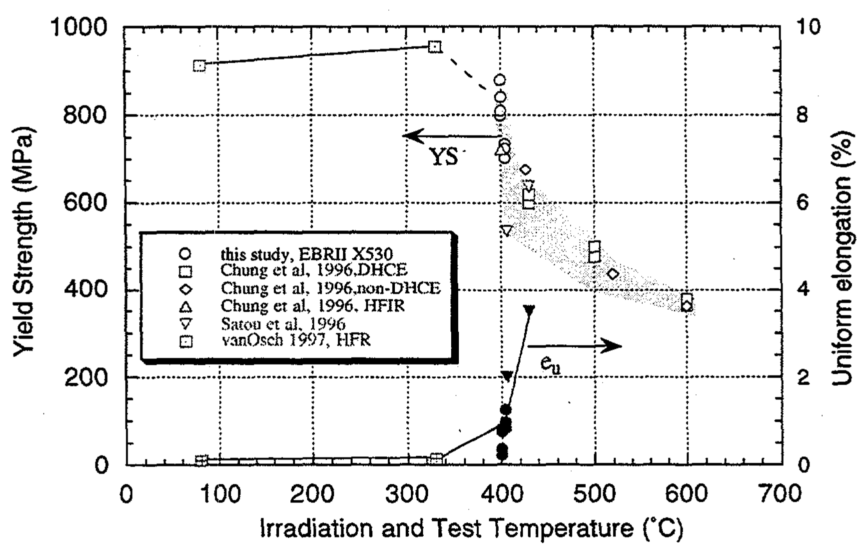

Fig. 2. Yield strength (open symbols) and uniform elongation (filled symbols) of V-(4-5)Cr-(4-5)Ti alloys irradiated and tested at temperatures between 80 and 600°C [7,9,14,15]. The damage levels were 4-6 dpa in ref. [15], 10-31 dpa in refs. [7,14], and 24-49 dpa in ref. [9]. The tensile elongations from refs. [7,14] have not been plotted, pending re-evaluation by ANL [8].

It is interesting to note that the V-5Cr-5Ti alloy had slightly better irradiated tensile elongations than the V-4Cr-4Ti alloy. As shown in Fig. 2, the yield strength and uniform elongation of irradiated V-(4-5)Cr-(4-5)Ti alloys appear to be strongly dependent on irradiation temperature between ~330 and 430°C [7,9,14,15]. The V-4Cr-4Ti and V-5Cr-5Ti tensile specimens were irradiated in different subcapsules in the EBR-II X530 experiment, which produced a slightly higher irradiation temperature (~400 vs. ~405°C) and lower damage level (~3.6 vs. ~4 dpa) in the V-5Cr-%Ti specimens compared to the V-4Cr-4Ti specimens. Further work is needed to determine the effect of damage level, composition and heat-to-heat variations on the radiation hardening behavior of vanadium alloys in this temperature regime.

The initial Charpy impact testing was conducted on specimens in the as-irradiated condition. The results are shown in Table 4. All of the specimens showed very low energies for fracture at all test temperatures (20-300°C), with the exception of the V-5Cr-5Ti (BL63) material, which showed a moderate energy absorption. The low energies measured for all of the specimens indicated that these specimens had become severely embrittled. To verify that this was an irradiation effect and not one caused by pickup of an embrittling impurity such as hydrogen during the irradiation or capsule disassembly, four specimens were annealed for 1 hour at 400°C in vacuum (~2×10^{-6} torr). The Charpy impact results for these specimens are also given in Table 4. The postirradiation-annealed specimens showed similar low levels of energy absorption as the as-irradiated specimens, confirming that the brittle behavior was not caused by the absorption of an interstitial impurity such as hydrogen.

The only specimens which showed some energy absorption were from the V-5Cr-5Ti alloy heat treated at 950°C. The load-displacement traces captured during the fracture process did not show a sudden load drop that would indicate cleavage fracture, despite the low values of energy absorbed. This suggests that the fracture process took place by the relatively slow growth of a crack across the specimen, rather than rapid crack extension by cleavage fracture. Scanning electron microscopy of the fracture surfaces showed transgranular cleavage for the V-4Cr-4Ti specimens and the V-5Cr-5Ti TWCA anneal specimens. The V-5Cr-5Ti specimens that were given a preirradiation anneal at 1050°C showed a mixture of transgranular cleavage and intergranular failure. The CR/950 V-5Cr-5Ti specimens exhibited a mixture of transgranular cleavage and ductile tearing (with intermixed laminated cracking, possibly associated with a precipitate band structure). It should be noted that postirradiation annealing at 400°C of V-5Cr-5Ti CR/950 specimens did not result in any change in the energy absorption (Table 4), again indicating that pickup of hydrogen was not responsible for the observed embrittlement.

Table 4. Summary of Charpy impact tests on EBR-II X530 specimens

Alloy	Preirradiation heat treatment	Spec. No.	Condition	Test Temperature	Absorbed energy (J)
V-4Cr-4Ti	TWCA&950	71-01	as-irradiated	100°C	0.1
"	"	71-02	as-irradiated	200°C	0.0
"	"	71-03	as-irradiated	285°C	0.4
"	"	71-04	vacuum anneal	200°C	0.4
"	"	71-05	vacuum anneal	275°C	0.4
V-5Cr-5Ti	TWCA ann.	63-1	as-irradiated	100°C	0.0
"	"	63-2	as-irradiated	200°C	0.1
"	"	63-3	as-irradiated	285°C	0.4
"	CR/950	63CR9-01	as-irradiated	200°C	2.0
"	"	63CR9-02	as-irradiated	285°C	2.8
"	"	63CR9-03	vacuum anneal	200°C	2.2
"	"	63CR9-04	vacuum anneal	275°C	2.8
"	CR/1050	63CR10-01	as-irradiated	200°C	0.7
"	"	63CR10-02	as-irradiated	285°C	0.4

ACKNOWLEDGEMENTS

The authors thank H. M. Chung and D. L. Smith (ANL) for their assistance with the specimen fabrication and irradiation, and A. M. Williams (ORNL) for providing the SEM analysis.

REFERENCES

1. D.L. Smith, H.M. Chung, B.A. Loomis and H.-C. Tsai, J. Nucl. Mater. 233-237 (1996) 356.
2. H. Matsui et al., J. Nucl. Mater. 233-237 (1996) 92.
3. D.J. Alexander et al., in Fusion Materials Semiannual Progress Report for Period ending June 30, 1996, DOE/ER-0313/20 (Oak Ridge National Lab, 1996) p. 87.
4. B.A. Loomis, L.J. Nowicki and D.L. Smith, J. Nucl. Mater. 212-215 (1994) 790.
5. B.A. Loomis, H.M. Chung, L.J. Nowicki and D.L. Smith, J. Nucl. Mater. 212-215 (1994) 799.
6. H.M. Chung, B.A. Loomis and D.L. Smith, J. Nucl. Mater. 212-215 (1994) 804.
7. H.M. Chung, B.A. Loomis and D.L. Smith, J. Nucl. Mater. 233-237 (1996) 466.
8. M.C. Billone, personal communication
9. M. Satou et al., J. Nucl. Mater. 233-237 (1996) 447.
10. M.L. Grossbeck, A.F. Rowcliffe and D.J. Alexander, in Fusion Materials Semiannual Progress Report for Period ending March 31, 1994, DOE/ER-0313/16 (Oak Ridge National Lab, 1994) p. 244.
11. H. Tsai et al., in Fusion Materials Semiannual Progress Report for Period ending September 30, 1994, DOE/ER-0313/17 (Oak Ridge National Lab, 1994) p. 8.
12. H. Tsai et al., in Fusion Materials Semiannual Progress Report for Period ending March 31, 1995, DOE/ER-0313/18 (Oak Ridge National Lab, 1995) p. 85.
13. S.J. Zinkle, A.N. Gubbi and W.S. Eatherly, in Fusion Materials Semiannual Progress Report for Period ending December 31, 1996, DOE/ER-0313/21 (Oak Ridge National Lab, 1996) in press.
14. H.M. Chung, L. Nowicki and D.L. Smith, in Fusion Materials Semiannual Progress Report for Period ending June 30, 1996, DOE/ER-0313/20 (Oak Ridge National Lab, 1996) p. 84.
15. E. van Osch, personal communication.