ASSESSMENT OF THE RADIATION-INDUCED LOSS OF DUCTILITY IN V-Cr-Ti ALLOYS — A. F. Rowcliffe and S. J. Zinkle (Oak Ridge National Laboratory)

OBJECTIVE

To assess the effects of neutron irradiation on the tensile and Charpy impact behavior of alloys in the V-Cr-Ti system.

SUMMARY

The current status of the irradiation data base on V-(4-5%Cr)-(4-5%Ti) alloys for tensile and Charpy impact properties is reviewed. Possible factors contributing to major inconsistencies in the data are examined.

INTRODUCTION

Alloys based on the V-Cr-Ti system are attractive candidates for structural applications in fusion systems because of their low activation properties, high thermal stress factor (high thermal conductivity, moderate strength, and low coefficient of thermal expansion), and their good compatibility with liquid lithium. The U.S. program has defined a V-4Cr-4Ti (wt %) alloy as a leading candidate alloy based upon evidence from laboratory-scale (30 kg) heats covering the approximate composition range 0-8 wt % Ti and 5 to 15 wt % Cr. A review of the effects of neutron displacement damage, helium, and hydrogen generation on mechanical behavior, and of compatibility with lithium, water, and helium environments was presented at the ICFRM-5 conference at Clearwater in 1991 [1]. The results of subsequent optimization studies, focusing on the effects of fast reactor irradiation on tensile and impact properties of a range of alloys, were presented at the ICFRM-6 conference at Stresa in 1993 [2,3]. The primary conclusion of this work was that the V-4Cr-4Ti alloy composition possessed a near-optimal combination of physical and mechanical properties for fusion structural applications. Subsequently, a production-scale (500 kg) heat of V-4Cr-4Ti (Heat No. 832665) was procured from Teledyne Wah-Chang [4], together with several 15 kg heats of alloys with small variations in Cr and Ti.

Further measurements of the effects of neutron irradiation on the swelling and mechanical properties of a range of alloy compositions were carried out in the Dynamic Helium Charging Experiment (DHCE) in FFTF. Neutron doses reached 31 dpa at 425°C and 18 dpa at 525 and 600°C; helium was generated at rates between 0.1 to 4.2 appm/dpa. It was reported that for a laboratory heat of V-4Cr-4Ti (designation BL 47) and several other alloys, uniform elongation values remained in excess of 8% for testing temperatures over the range 25 to 600°C. These data were presented in papers at the ICFRM-7 conference at Obninsk [5,6]. Additional tensile data from the DHCE experiment were presented recently from which it was concluded that the tensile ductilities of V-8Cr-6Ti and V-9Cr-6Ti also remained in excess of 7% uniform and 11% total elongation for test temperatures in the range 25 to 600°C [7].

The favorable mechanical properties data and promising irradiation performance of the V-4Cr-4Ti composition encouraged the ITER project in 1993 to consider this alloy for the first wall/shield structure and other high heat flux applications. To establish an irradiation performance data base relevant to ITER conditions, a series of reactor experiments were initiated by the U.S. program to explore the effects of neutron irradiation at temperatures below the lowest temperature (430°C) attained in the FFTF experiments. These experiments included HFBR V1-V4 (0.1, 0.4 dpa; 110 to 500°C), BOR-60 Fusion 1 (12 dpa, 315-340°C), ATR (5 dpa; 200, 300°C), EBR-II X530 (4 dpa, 370-410°C), and HFIR 400 J (8 dpa, 400°C). The primary focus of these experiments was on the 500 kg production heat of V-4Cr-4Ti. Initial tensile results from the experiments in HFBR and EBR-II indicate that neutron irradiation at temperatures up to 400°C induces large increases in yield stress with a concomitant major reduction in strain hardening capacity and uniform elongation [8, 9]. These changes in deformation behavior are coupled with large increases in the ductile-to-brittle transition temperature measured in 1/3-size Charpy impact tests. The tensile and impact property data reported for the irradiation experiments carried out at temperatures up to 400°C are not consistent with the earlier data reported for irradiation at 430°C to 600°C; possible sources of these inconsistencies are explored in the following.

Measurement of Uniform Elongation

The uniform elongation is a measure of the permanent plastic strain that accumulates prior to necking and failure in a uniaxial tensile test. The ability of a material to strain-harden and the extent of the uniform elongation are extremely important factors in the ability of a material to accommodate secondary stresses. The uniform elongation is utilized in component design to develop design rules to guard against various types of failure [10]. When ε_u exceeds 5%, a material is considered to be ductile and the ASME Code rules are applicable. For ε_u values in the range 1 to 5%, materials are considered to be semi-brittle and additional design rules are required; for example, bending stresses have to be reduced to guard against the embrittlement of outer fibers. In situations where ε_u is reduced to <1%, a material is considered to exhibit brittle behavior with enhanced sensitivity to the presence of flaws and notches; design rules based upon fracture mechanics principles become more appropriate in this situation.

In a tensile test, the region of uniform plastic strain occurs after yielding when the gage length extends uniformly together with a uniform reduction in cross-sectional area. Eventually, the strain-hardening capacity is exhausted; further localized reductions in area and increase in local stress are no longer balanced by an increase in material strength. Beyond this point of maximum load, further plastic deformation is localized in the necked region. Uniform engineering strain in a tensile test is the plastic elongation at the point of maximum load. Contributions to the overall extension from the elastic extension of the specimen are generally subtracted from the reported elongations, as described in the ASTM Standard Test Method for Tension Testing of Metallic Materials (E345-87, paragraph 7.6.6.1). More importantly, elastic extensions associated with the specimen gripping system must be excluded. This component is automatically excluded when the specimen gage extension is measured with an extensometer, and must be excluded by graphical analysis when specimen elongation is monitored from the cross-head displacement. Fig. 1 compares room temperature engineering stress-displacement curves for V-4Cr-4Ti determined by extensometry and by cross-head displacement measurement on the same specimen. In the extensometry case, the strain that precedes yielding is ~0.3% and consists entirely of elastic deformation of the specimen. However, when the cross-head movement is measured there is an effective displacement of ~3.0% before yielding which consists mainly of elastic strains in the specimen gripping system. For irradiated materials with low strain-hardening rates, inclusion of this system displacement can produce a large over-estimate of uniform strain.

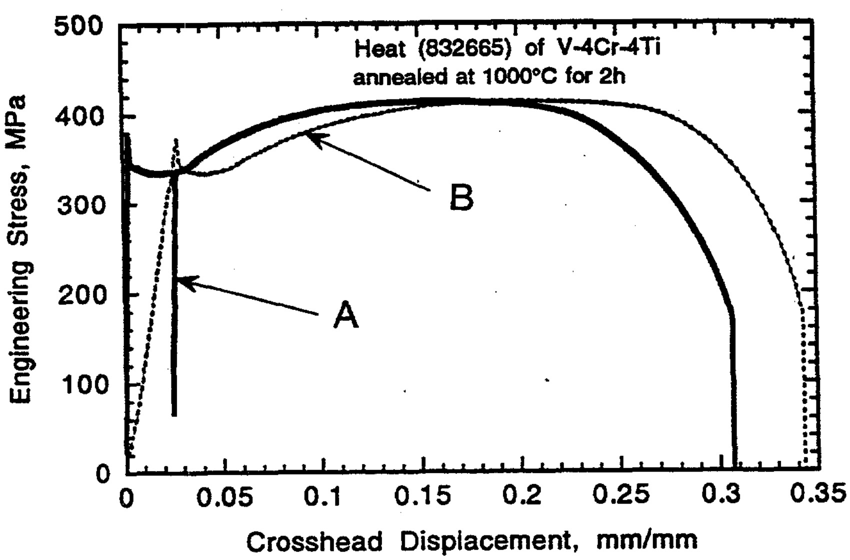

Fig. 1. Comparison of engineering stress-displacement curves for V-4Cr-4Ti determined by extensometry (A) and by crosshead-movement (B).

Tensile Data for Irradiated V-Cr-Ti Alloys

Data that have been reported for V-(4-5)Cr-(4-5)Ti alloys irradiated in various reactors are summarized in Table 1. The earlier data reported for FFTF/MOTA experiments are for small experimental heats; more recently data have been obtained for the production heat (832665) following irradiation to low doses at temperatures ≤420°C. Yield strength at the irradiation temperature is plotted as a function of irradiation temperature in Fig. 2. These data are from a variety of sources; the data for the HFR irradiation are from unpublished work by van Osch et al., Netherlands Energy Research Foundation ECN, Petten. For irradiation temperatures below 400°C, the irradiated yield stress is fairly independent of temperature, reaching values of ~650 MPa at 0.4 dpa and increasing to 900-950 MPa for doses of 4 to 6 dpa. Radiation hardening decreases fairly rapidly with increasing temperatures over the range 400 to 450°C and then decreases more gradually up to 600°C. At 600°C (0.32 Tm), radiation hardening at ~35 dpa is still significant, ($\Delta\sigma_y$, ~130 MPa).

From the limited data shown in Fig. 3, it appears that ε_u begins to increase with increasing irradiation above ~400°C commensurate with the strong decrease in radiation hardening shown in Fig. 2. The ε_u values reported for the FFTF/MOTA experiments at 425-600°C have not been included in Fig. 3. Following extensive discussions within the U.S. program, it now appears that these data have not been reported correctly. ANL researchers are currently re-evaluating the irradiated tensile data for a range of alloys [13], and the corrected values will be published in the near future.

Charpy Impact Data for V-Cr-Ti Alloys

In addition to the discrepancies regarding the effects of neutron irradiation on uniform elongation at temperatures >400°C, there are several conflicting aspects of the reported CVN impact data for the V-4Cr-4Ti alloy. The U.S. vanadium alloy program, following the ferritic-martensitic steels program, has adopted the one-third-size Charpy vee-notch specimen (CVN) as a convenient means of assessing changes in fracture behavior induced by heat-treatment or by irradiation. It has been pointed out by G. R. Odette et. al. that this specimen has some serious shortcomings and that caution must be exercised in interpreting results; not only does the specimen lack the capacity to provide valid intrinsic properties, but it may even fail to detect the propensity for a material to fail by cleavage [16].

Data are available from three separate irradiation experiments designed to assess the effect of neutron irradiation on the impact behavior of V-4Cr-4Ti. Specimens from the experimental heat (BL 47) were irradiated in the FFTF/MOTA (cycles 9-11) experiment at 425, 520, and 600°C to 24 to 36 dpa [3]. Specimens fabricated from heat 832665 were irradiated at lower temperatures in two separate experiments, and data have been reported for specimens irradiated to 0.4 dpa in HFBR at 100, 200, and 230°C [8]. In this current volume of the Semiannual Report, data are also reported on heat 8332665, following irradiation at 380-405°C to ~4 dpa in EBR-II [9].

The CVN data reported for the FFTF/MOTA (cycles 9-11) irradiation experiment at 425°C indicated that the heat BL 47 was highly resistant to cleavage failure following irradiation. However, the other two sets of data for V-4Cr-4Ti from the HFBR and EBR-II experiments are in complete contrast to these data. Large upward shifts in DBTT were observed for specimens irradiated over the range 110 to 400°C; in particular, the specimens of the production heat irradiated in EBR-II at 400°C exhibited brittle cleavage behavior in impact tests carried out at temperatures as high as ~280°C. Using one-half of the upper shelf energy as an index, the radiation-induced increase in DBTT is plotted in Fig. 4 against the corresponding increase in the yield stress at the DBTT (the latter were obtained by interpolation between tensile tests carried out at RT and at the irradiation temperature). Also shown are the FFTF/MOTA data reported for the experimental heats V-3Ti-1Si (BL 45), V-5Ti (BL 46), V-18Ti (BL 15), V-14Cr-5Ti (BL 24), and V-8Cr-6Ti (BL 49). With the striking exception of alloys BL 45, BL 46, and BL 47, the data fit reasonably well to a linear relationship between DBTT shift and radiation hardening with a slope of ~0.7°C/MPa. Similar relationships occur for other irradiated materials exhibiting stress-controlled fracture, such as pressure vessel steels and ferritic-martensitic steels [17]. However, the data for alloys BL 45, 46, and 47 do not follow this relationship. It was reported that CVN specimens for all three alloys bent without fracture at the lowest test temperature, i.e. -196°C; DBTTs of ~-200°C were attributed to each of these alloys.

Table 1. Tensile Properties of V-(4-5)Cr-(4-5)Ti Alloys Irradiated in Various Reactors

Alloy	Irradiation Experiment	Tirr (°C)	Dose (dpa)	σ_y	σ_u	ε_u	ε_t	Ref.
4Cr-4Ti BL47	FFTF/MOTA (cycles 9-11)	420 520 600	~34 ~28 ~27	600 460 365	700 545 460	8 10 13.5	10 14 19	[2]
4Cr-4Ti BL47	FFTF/MOTA 2B (DHCE)	430 500 600	18-31 18-31 18-31	600 410 365	700 570 450	10 9.5 13.5	12.5 14 19	[12]
4Cr-4Ti BL47	HFIR RB* -400 J1	400	8	710	800	4.5	12	[11]
5Cr-5Ti	FFTF/MOTA 2B (DHCE)	430	24	637	714	3.5	8.1	[14]
5Cr-5Ti	BR-2	100	0.7	785	—	—	3	[15]
5Cr-5Ti	FFTF/MOTA (2A)	407	49	535	613	2.0	5.8	[14]
5Cr-5Ti BL63	EBR-II X-530	405	3.6	719	761	1.0	5.7	[9]
4Cr-4Ti 832665	EBR-II X-530	400	4	832	849	0.5	5.0	[9]
4Cr-4Ti 832665	HFBR V1, V2	110 205 240	0.4 0.4 0.4	655 647 637	655 647 637	0.1 0.1 0.1	9.7 9.2 9.0	[8]

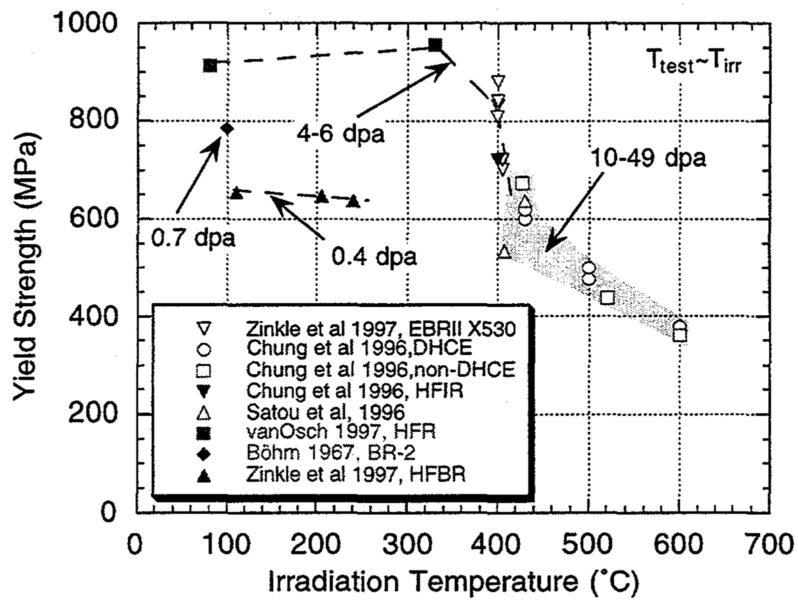

Fig. 2. Temperature dependence of yield strength measured at the irradiation temperature for alloys irradiated in various reactors.

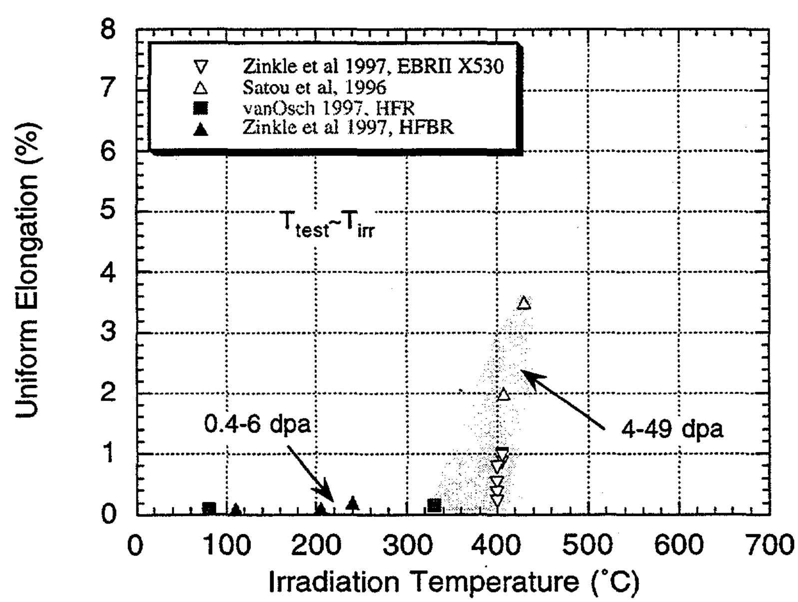

Fig. 3. Temperature dependence of uniform elongation measured at the irradiation temperature for alloys irradiated in various reactors. Data from ANL studies are not presented, pending re-evaluation of published values.

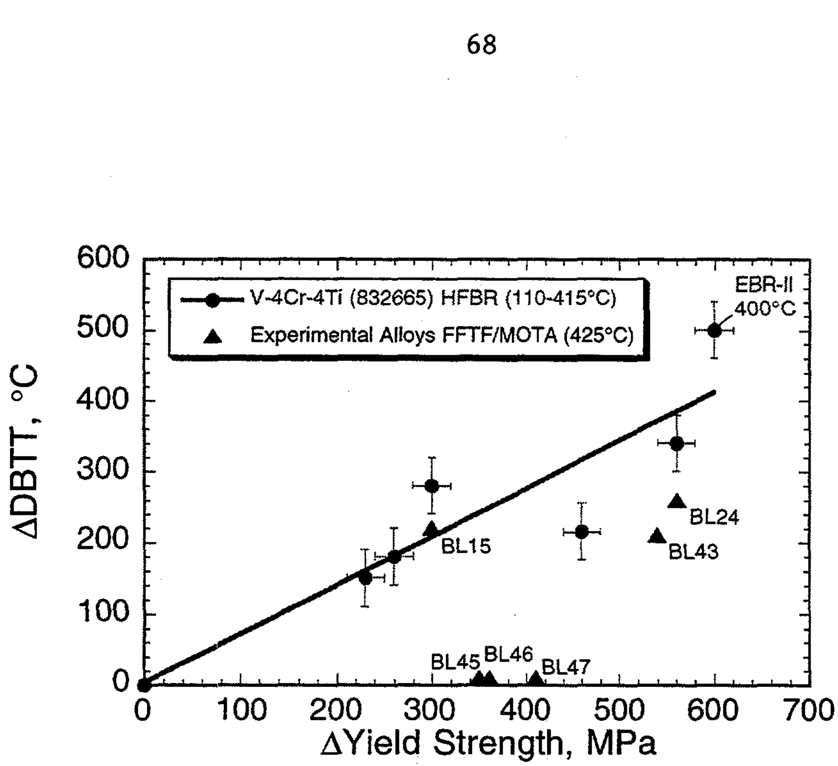

Fig. 4. Increase in DBTT versus increase in room temperature yield stress for heat 832665 irradiated in HFBR (110, 200, 235, 315, and 420°C) and in EBR-II (400°C), and for heat BL 47 irradiated in FFTF/MOTA at 425°C.

The failure to initiate cleavage fracture in these three alloys is surprising in view of the magnitude of the radiation-induced increases in room temperature (RT) yield strength (340-410 MPa). The published absorbed energy vs. test temperature curves for the V-4Cr-4Ti (BL 47) and V-5Ti (BL 46) [3] exhibit unusual behavior in that the absorbed energy initially decreased with decreasing test temperature, reached a minimum at around -120°C and then increased back to upper shelf values at liquid nitrogen temperature. The possibility that this behavior is somehow related to hydrogen diffusion seems implausible since (a) the specimens were vacuum annealed at 400°C prior to testing, and (b) unirradiated specimens charged with hydrogen in the same experiment do not show this behavior.

The three alloys V-5Ti, V-3Cr-1Si, and V-4Cr-4Ti have been reported as being "virtually immune to neutron displacement damage," [18]. Since these alloys are not immune to radiation hardening, their exceptional behavior must derive from an intrinsic resistance to the initiation of cleavage fracture. Considering only the V-4Cr-4Ti composition, there appear to be major differences in the fracture behavior of the two heats BL 47 and 832665. Following irradiation in FFTF/MOTA at 425°C, the BL 47 heat reportedly undergoes plastic collapse rather than cleavage fracture in CVN tests at all temperatures, even though the irradiated RT yield stress was ~800 MPa. On the other hand, following irradiation in HFBR at 275°C, the production heat exhibited brittle cleavage fracture in CVN tests at temperatures <150°C when the irradiated RT yield stress was only ~620 MPa. The production heat (832665) is a scaled-up version of BL 47 and the only significant difference in composition is the lower levels of nitrogen and carbon (~80 wppm vs. ~200 wppm) for the production heat.

Taken at face value, the Charpy test results on unirradiated specimens suggest that the two heats of V-4Cr-4Ti differ in their response to heat treatment. In Fig. 5, data are presented on the effects of recrystallization temperature on the impact properties of the large heat. The ANL data illustrate that in a partially recrystallized condition, produced by annealing 50% CW material at 950°C for 1 hr, the production heat does not exhibit a transition temperature [19]. However, ORNL data show that a fully recrystallized microstructure (1050°C, 2 hrs) exhibits a transition at ~-180°C [20], and this is increased to -140°C when the recrystallization temperature is increased to 1100°C. This type of behavior has been observed in other V alloys. Figure 6 illustrates the effect of recrystallization temperature on a V-5Cr-5Ti alloy (designated BL 63) [21]. The ORNL data, demonstrating an increase in the DBTT with increasing recrystallization temperature for the small heat of V-5Cr-5Ti (BL 63), were obtained using a CVN specimen that was machined in an L-T orientation with a machined notch depth of 0.5 mm, a notch root radius of 0.08 mm,

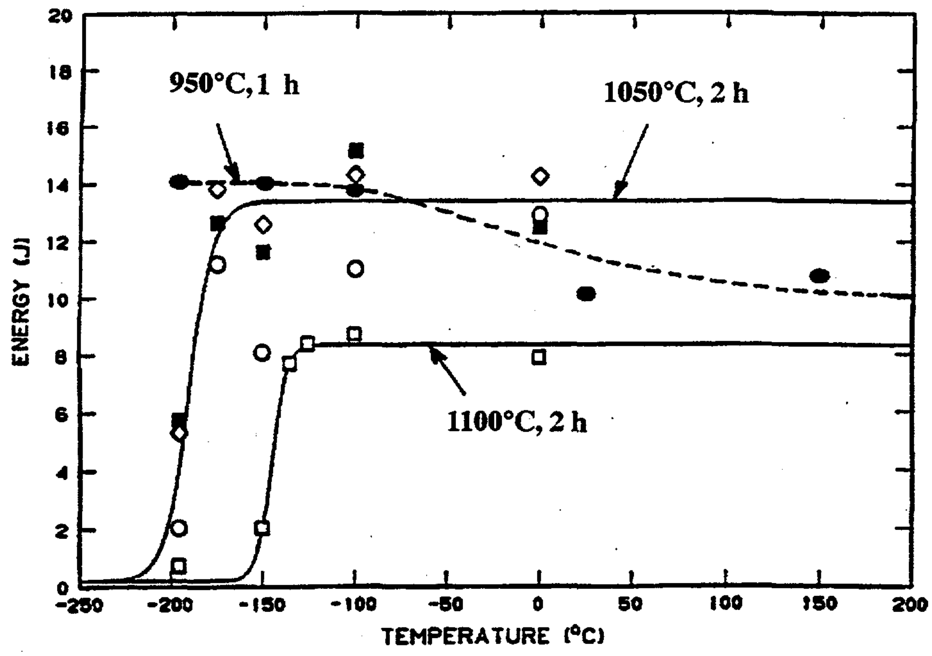

Fig. 5. Unirradiated Charpy impact properties for heat 832665 following recrystallization treatments at 950°C for 1 hr [18], 1050°C for 2 h [19], and 1100°C for 2 h [ORNL data].

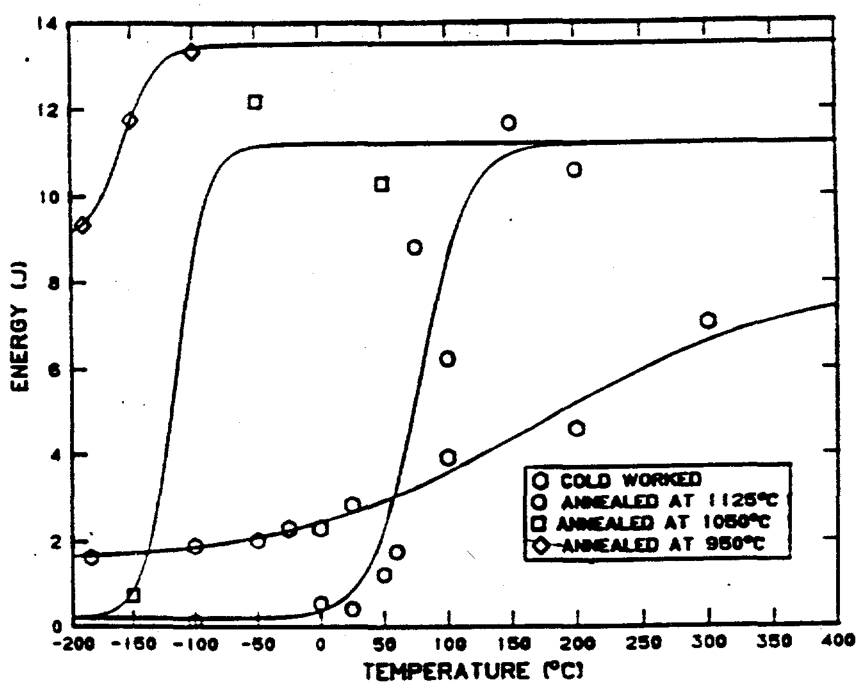

Fig. 6. Effect of recrystallization temperature on the Charpy impact properties of a V-5Cr-5Ti alloy, heat BL 63 [20].

and a notch angle of 30° [21]. For the work on the production heat, the notch depth was increased to 0.67 mm. Other examples of the dependence of DBTT on microstructure have been presented for a V-6Cr-3Ti alloy [19] and a V-4Cr-4Ti-0.1 Si alloy [22]. The BL 47 heat of V-4Cr-4Ti reportedly does not follow the same type of behavior. H. M. Chung et al., [23] reported that heat BL 47 did not a exhibit transition in impact behavior following annealing treatments at 1050 or 1125°C, when the material was presumably in a fully recrystallized condition (Fig. 7). In the same study, both V-5Cr-5Ti and V-5Cr-3Ti exhibited cleavage fracture between -50 and -100°C; the superior behavior of heat BL 47 was ascribed to the absence of various types of precipitation related to impurities of Cl, S, P, Cu, Ne, and K in the other two alloys. While it is possible that the BL 47 heat does indeed possess some undefined combination of impurity and microstructural characteristics that confer superior fracture resistance relative to the production heat, it is equally possible that extrinsic factors relating to the testing methodology are playing a significant role. Although the details of notch geometry for the CVN specimens used in Ref. [23] were not described, ANL workers have reported using an L-S orientation and a machined notch with 0.61 mm depth, 0.25 mm root radius, and a 45° notch angle in other studies on heat BL 47 [24]. The ORNL studies on the large heat of V-4Cr-4Ti used an L-T orientation and a 30° machined notch with 0.67 mm depth and 0.08 mm root radius [8]. Given the marginal capacity of the 1/3-size CVN specimen [16], it is entirely plausible that these differences in notch geometry could account for the appearance of transition behavior in V-4Cr-4Ti in some studies but not in others.

Although arguments based on intrinsic fracture resistance cannot be completely ruled out, explanations for the anomalous irradiation behavior of alloys BL 45, 46, and 47 based upon extrinsic experimental factors would appear to be entirely credible and require further consideration as follows.

(a) It has been shown by Odette et al. [16], that for the production heat of V-4Cr-4Ti, the transition from ductile behavior to cleavage fracture occurs over a fairly narrow temperature range, and that the transition temperature decreases with decreasing strain and with decreasing degree of constraint. They conclude that for the unirradiated alloy, blunt-notched MCVN specimens may not have the capacity to initiate cleavage fracture. These observations raise the possibility that depending upon the selection of notch orientation (L-S, L-T) and notch geometry (depth, angle, radius), the MCVN specimens used for the testing of the BL 45, 46, and 47 alloys did not have the capacity to initiate cleavage even in a radiation-hardened condition. Experimental details regarding notch orientation and geometry for these specimens have not been published.

(b) The CVN impact specimens from the FFTF/MOTA (cycles 9-11) experiments were reportedly given a postirradiation anneal at 400°C for 1 hour in a vacuum to remove hydrogen prior to testing. If the specimen annealing temperatures were actually higher than 400°C, then some of the radiation damage could have annealed out prior to testing. This could conceivably have more of an effect on the alloys in the experiment with the least amount of radiation hardening (BL 45, 46, 47). This could be investigated by verification of the annealing temperature measurement system and by hardness measurements on broken specimens.

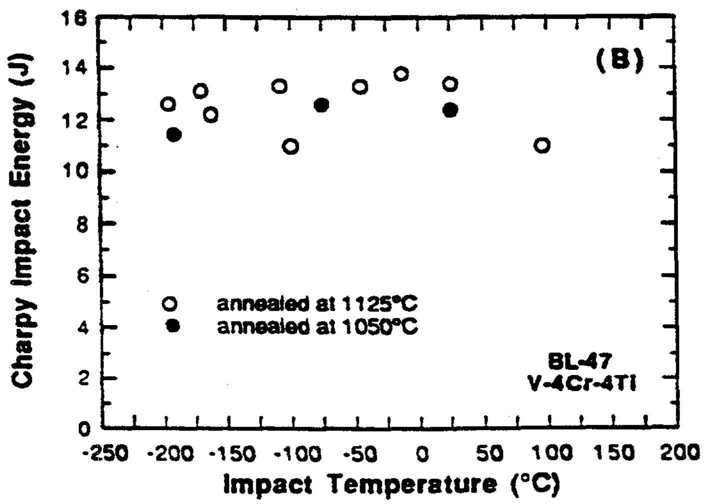

Fig. 7. Effect of annealing temperature on the Charpy impact properties of V-4Cr-4Ti, heat BL 47 [23].

CONCLUSIONS

Severe radiation hardening occurs for V-(4-5%)Cr-(4-5%)Ti alloys for irradiation temperatures up to ~400°C; a sharp decrease in hardening over the range 400-450°C is followed by a more gradual decrease up to 600°C. For irradiation temperatures ≤405°C, hardening is accompanied by a severe reduction in strain-hardening capacity and uniform elongation. The effects of irradiation at higher temperature on tensile ductility must be regarded as uncertain pending a re-evaluation of the ANL data.

There are major inconsistencies in the data reported for the effects of neutron irradiation on CVN impact properties. Earlier ANL data reported for experimental heats of V-5Ti, V-3Ti-1Si, and V-4Cr-4Ti are incompatible with the ΔDBTT versus $\Delta\sigma_y$ relationship determined for the production heat of V-4Cr-4Ti and several other experimental alloys. Explanations of this behavior based upon a combination of extrinsic experimental factors are more credible than arguments based upon an intrinsic resistance to cleavage fracture.

As recommended by Odette et al. [16], the effects of constraint and loading rate must be accounted for in future studies on the effects of compositional and microstructural variants and radiation damage on the fracture behavior of vanadium alloys.

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