Available online at www.sciencedirect.com
ScienceDirect
Journal of Nuclear Materials 367–370 (2007) 637–643
journal of nuclear materials
www.elsevier.com/locate/jnucmat

Brittle–ductile transitions in vanadium and iron–chromium

T.D. Joseph, M. Tanaka, A.J. Wilkinson, S.G. Roberts *

Department of Materials, University of Oxford, Oxford OX1 3PH, UK

Abstract

We report ongoing experimental work on fracture and brittle–ductile transitions in vanadium, iron and iron–9% chromium single crystals and polycrystals. Tests were carried out by four-point bending of pre-cracked specimens in the temperature range 77–300 K, at a variety of strain rates. For a given material, variation of the brittle–ductile transition temperature with strain rate allows us to estimate an activation energy for the controlling process; for vanadium this was found to be ∼0.37 eV, for iron ∼0.21 eV and for iron–9% chromium ∼0.11 eV.

1. Introduction

Materials currently anticipated for pressure-vessel structural components of the proposed DEMO prototype fusion power plant include reduced activation ferritic–martensitic (RAFM) steels based on iron–9% chromium [1,2], though vanadium alloys [2] and tungsten [3] have also been suggested for some specialist applications. These materials may offer adequate strength and toughness and minimal sensitivity to fast neutron/alpha particle irradiation effects such as swelling and increase in brittle–ductile transition temperature. However, it has been noted that for RAFM steels, 'Further reduction of brittle–ductile transition temperatures should be pursued' [4]. It is difficult, expensive and time-consuming to perform experiments on irradiated materials. Modelling of effects of irradiation on flow properties is now becoming more frequent (e.g. [5,6]). While it is 'obvious' that easier plastic flow implies a less brittle material,[1] explicit links between flow behavior and fracture are less easy to model, except, at present, for relatively simple, generally single crystal, materials.

Many crystalline solids fail by brittle cleavage at low-temperatures, and by plastic processes at high temperatures. For most materials, the fracture stress intensity factor (K_{Ic}) increases from 'lower shelf' values associated with brittle fracture over a brittle–ductile-transition temperature range of 100 K or more: single crystal BCC metals [7–9], intermetallics [10–12], MgO [13], germanium [14] and most other materials exhibit this type of transition (as of course do polycrystalline ferritic steels [15]). In

* Corresponding author.
E-mail address: steve.roberts@materials.ox.ac.uk (S.G. Roberts).

[1] 'Obvious', but not always true, as in materials with limited plasticity, stresses resulting from dislocation pile-ups at grain boundaries, second-phase particles or locked dislocations can initiate cleavage.

0022-3115/$ - see front matter © 2007 Published by Elsevier B.V.
doi:10.1016/j.jnucmat.2007.03.077

some cases, e.g. for Si [16,17] and Al_2O_3 (sapphire) single crystals [18], the transition is very sharp, occurring over a temperature range of <10 K. The transitions are strain-rate dependent. For BCC metals the strain-rate dependence is small; for Si single crystals, increasing the strain rate by a factor of ten increases the temperature of the transition typically by 100 K. The strain-rate variation of the transition temperature T_c allows an activation energy E_a to be calculated:

$$\frac{d\varepsilon}{dt} = A\exp\left[\frac{-E_a}{kT_c}\right] \tag{1}$$

Where comparisons have been possible, this has been found to be equal to the activation energy for dislocation glide [19].

The increase in fracture toughness over the transition range is associated with an increase in the dislocation mobility resulting in an increasing amount of plasticity around the crack tip. A plastic zone is formed, which reduces the high stresses in front of the crack tip through elastic interactions, and which also blunts the crack.

Models have been developed in which the developing crack-tip plastic zone is modelled as a self-organising array of dislocations emitted from one or more sources near to the crack tip; the dislocations move according to an experimentally-derived velocity/stress/temperature law [19–23]. The elastic ('shielding') interactions of the dislocations with the crack tip are calculated at each stage; when the total stress intensity at the crack tip reaches the low-temperature cleavage fracture toughness, fracture is assumed to occur. With increasing temperature, increasing dislocation activity gives increasing shielding of the crack tip, and thus increasing applied stress intensity for fracture.

Hence, for modelling brittle–ductile transitions in such materials, essential input data are: (a) a nil-ductility fracture toughness, and (b) dislocation velocity as a function of stress and temperature. The models' predictions then must be compared against (i) experimental data for fracture toughness as a function of temperature and strain rate, and, if possible, (ii) the sizes of plastic zones.

We describe here results of some initial fracture experiments for single crystal vanadium and polycrystalline iron and iron–9% chromium alloys, representing the major constituents of proposed structural alloys for fusion materials applications. They form a 'snapshot' of work presently in progress.

2. Experimental methods

Specimen materials – single crystal vanadium and polycrystalline iron and iron–9% chromium – were supplied by Metal Crystal and Oxides, Button End, Harston, Cambs, UK. Specimens were cut using a Buehler isomet saw and polished using a succession of diamond pastes, to a final 1 μm paste finish. Final specimen sizes were typically 10 mm long, with 800 μm square cross-section. Single crystal specimens were aligned so that each face was of {1 0 0} type. Polycrystals were etched to examine the grain structure; for all materials used, equiaxed grains of ~100–150 μm diameter were found. Notches, with associated fine pre-cracks, were made in the tensile faces of test specimens by electro-discharge machining. Specimens were tested to fracture over the temperature range 77–293 K, using four-point bending, with outer and inner contact spacings of 8 mm and 4 mm (V) and 10 mm and 4 mm (Fe, Fe–Cr).

3. Results

3.1. Single crystal vanadium

Fig. 1 shows bend test results for single crystal vanadium tested at a strain rates of $4.4 \times 10^{-5}\ s^{-1}$ and $4.1 \times 10^{-4}\ s^{-1}$. 'Brittle' means that specimens failed immediately at a critical load, with no plastic strain, giving a planar fracture surface; 'semi-brittle' means that specimens failed in a brittle manner but with yield and some plastic strain prior to fracture (again with a planar fracture surface), 'ductile' means that specimens bent with no fracture. For ductile specimens the fracture toughness value shown is that at which bending initiated, assuming a crack depth equal to the mean value for fractured specimens (i.e. slightly greater than the notch depth); the real fracture toughness under these conditions will be higher than this value. For a surface strain rate of $4.1 \times 10^{-4}\ s^{-1}$, the transition temperature, T_c, defined as the mean of the highest temperature at which brittle or semi-brittle fracture is observed and the lowest temperature at which ductile behavior is observed, is 164 K; for a strain rate of $4.3 \times 10^{-3}\ s^{-1}$, $T_c = 174$ K. For other strain rates tested, at $1.35 \times 10^{-2}\ s^{-1}$, $T_c = 186$ K, and at $4.4 \times 10^{-5}\ s^{-1}$, $T_c = 138$ K. If these results are interpreted using Eq. (1), the derived activation energy is 0.26 eV, somewhat lower than the activation energy for screw dislocation motion in vanadium of 0.55 eV found by Wang and Bainbridge [24].

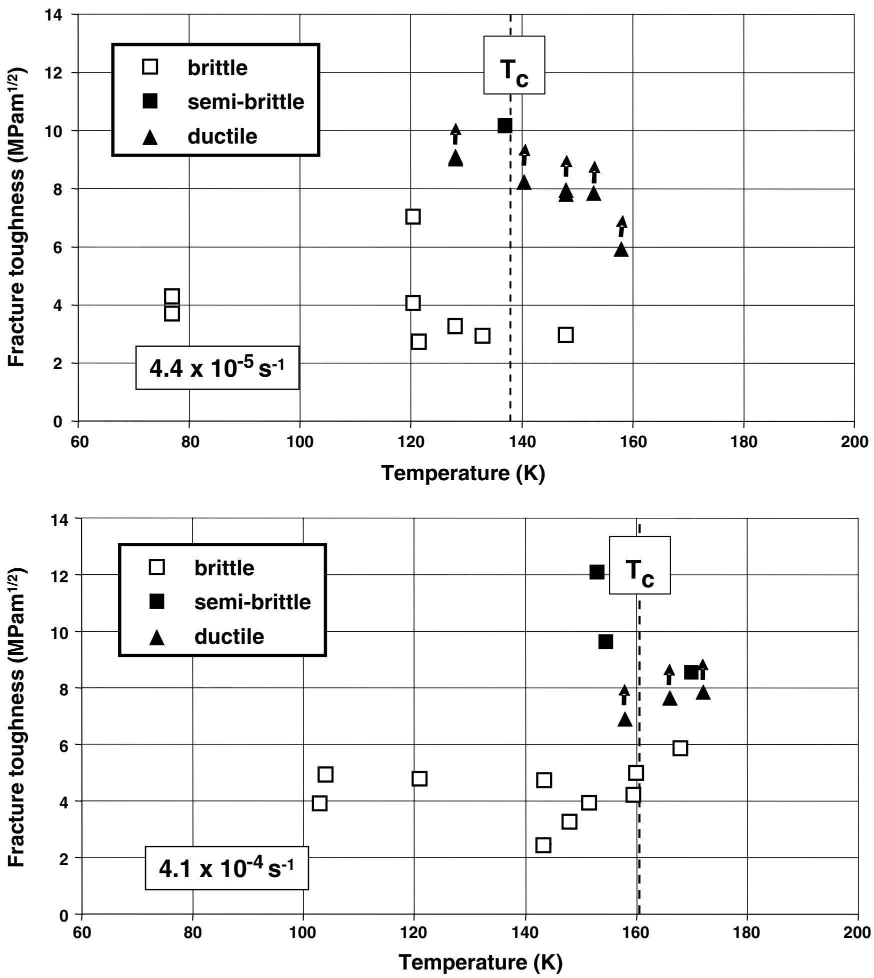

Fig. 1. Bend test results for single crystal vanadium at two strain rates.

For the brittle–ductile transitions shown in Fig. 1, there appears to be no systematic variation in fracture stress intensity below the transition temperature; however, within a narrow transition range, fracture can occur at elevated stress intensity, associated with a non-zero plastic strain at fracture. This is very different from, for example, molybdenum [7] and tungsten [9], where the stress to fracture was found to rise steadily over the full range between 77 K and T_c. Sharp transitions of the type observed here in vanadium have previously been associated only with dislocation-free covalently bonded materials [17,18], where dislocation activity at crack tip is triggered only just below the fracture load at T_c [20]. To determine the reasons for this unusual behavior in vanadium, dislocation activity around crack tips is presently being investigated by electron backscattered diffraction methods.

3.2. Iron and iron–9%chromium

Experiments have to date been preformed on polycrystalline materials only, as single crystals are currently unavailable with dimensions large enough to give the numbers of specimens required.

For polycrystalline iron, tests have been carried out at three strain rates: $4.46 \times 10^{-4}\ s^{-1}$, $8.93 \times 10^{-4}\ s^{-1}$ and $4.46 \times 10^{-3}\ s^{-1}$. The 77 K fracture toughness was found to be 20.6 ± 4 MPa m$^{1/2}$, with no systematic variation with strain rate. Stress–strain curves for tests at 77 K showed only elastic loading before fracture, as shown in Fig. 2(a)i, and produced fracture surfaces showing only transgranular cleavage, as shown in Fig. 2(b). For some tests at temperatures between 77 K and the brittle–ductile transition, fracture could similarly be identified as brittle. In the transition temperature range,

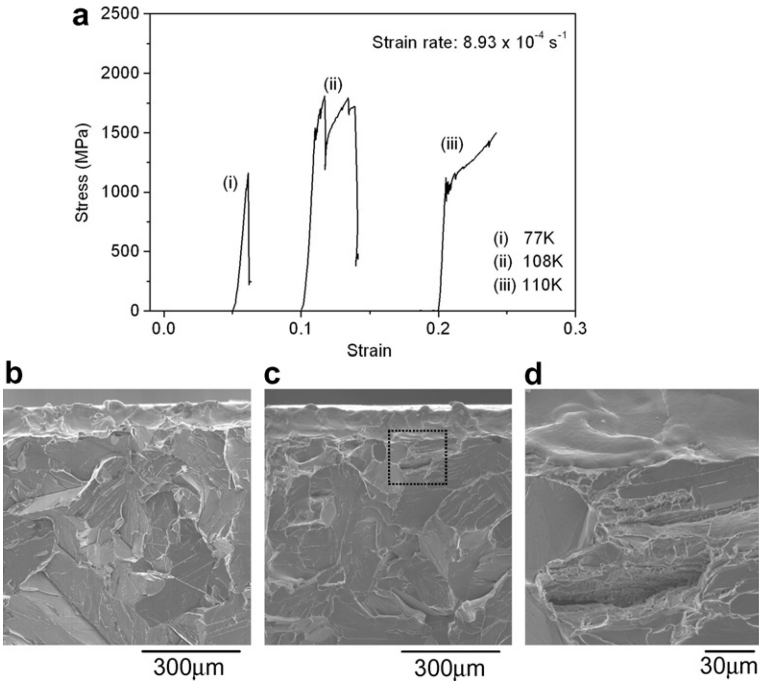

Fig. 2. Stress–strain curves from four-point bending tests of pure iron at a strain rate of $8.93 \times 10^{-4}\,s^{-1}$: (i) 77 K – brittle fracture, (ii) 108 K – semi-brittle fracture, (iii) 110 K – ductile bending; (b) fracture surface for 77 K test; (c) fracture surface for 108 K test; (c) enlargement of the area indicated in (c), showing voids around part of the pre-crack.

some stress–strain curves showed fracture after yield, as shown in Fig. 2(a)ii; the serrations on the stress–strain curves after yield were accompanied by audible clicks, which we tentatively associate with crack progression from one grain to the next. The fracture surfaces in these cases showed mostly transgranular cleavage (Fig. 2(c)), but with some evidence of local plastic flow near the crack tip (Fig. 2(d)). Tests showing these characteristics were identified as 'semi-brittle'. Above the brittle–ductile transition temperatures, specimens did not fracture, and stress–strain curves showed work-hardening after yield (Fig. 2(a)iii).

Some overlap was normally found between the semi-brittle and ductile temperature regimes, due to the variability in test results as a consequence of the large grain size compared to the test specimen size. The brittle–ductile transition temperature (T_c) was defined as the mean of the highest temperature at which a test showed brittle or semi-brittle behavior and the lowest temperature at which a test showed fully ductile behavior. Tests at the highest strain rate gave a brittle–ductile transition temperature of 114 K (±3 K). Fracture toughness values measured in the semi-brittle range (100–116 K) ranged from 22 to 31 MPa $m^{1/2}$. For the lower loading rates, transition temperatures were 103 K (±4 K) at $4.46 \times 10^{-4}\,s^{-1}$, and 106 K (±2 K) at $8.93 \times 10^{-4}\,s^{-1}$. Using Eq. (1), an activation energy for the BDT of ~0.21 eV is obtained, though given the temperature range and error margins of the T_c measurements, this can only be a tentative value.

For polycrystalline iron–9% chromium, tests have been carried out at the same three strain rates as for iron; see Fig. 3. The 77 K fracture toughness was found to be 17.6 ± 4 MPa $m^{1/2}$, with no systematic variation with strain rate. Iron–9 % chromium, like iron and unlike vanadium, has a brittle–ductile transition in which semi-brittle behavior is exhibited, where dislocation activity can raise the fracture toughness below T_c; however, the data do not show any clear temperature dependence of semi-brittle fracture toughness. Tests carried out at $4.46 \times 10^{-3}\,s^{-1}$ found a brittle–ductile transition between 127 K (cleavage) and 122 K (fully ductile), that is, about 10 K higher than for iron. For the tests at $4.46 \times 10^{-4}\,s^{-1}$, T_c was found to be 101 K (±1 K), and at $8.93 \times 10^{-4}\,s^{-1}$, T_c was found to be 107 K (±2 K). If these results are interpreted using Eq. (1), the derived activation energy is ~0.11 eV.

Fig. 4 shows the side faces near the notch root of iron–9% chromium polycrystalline specimens tested

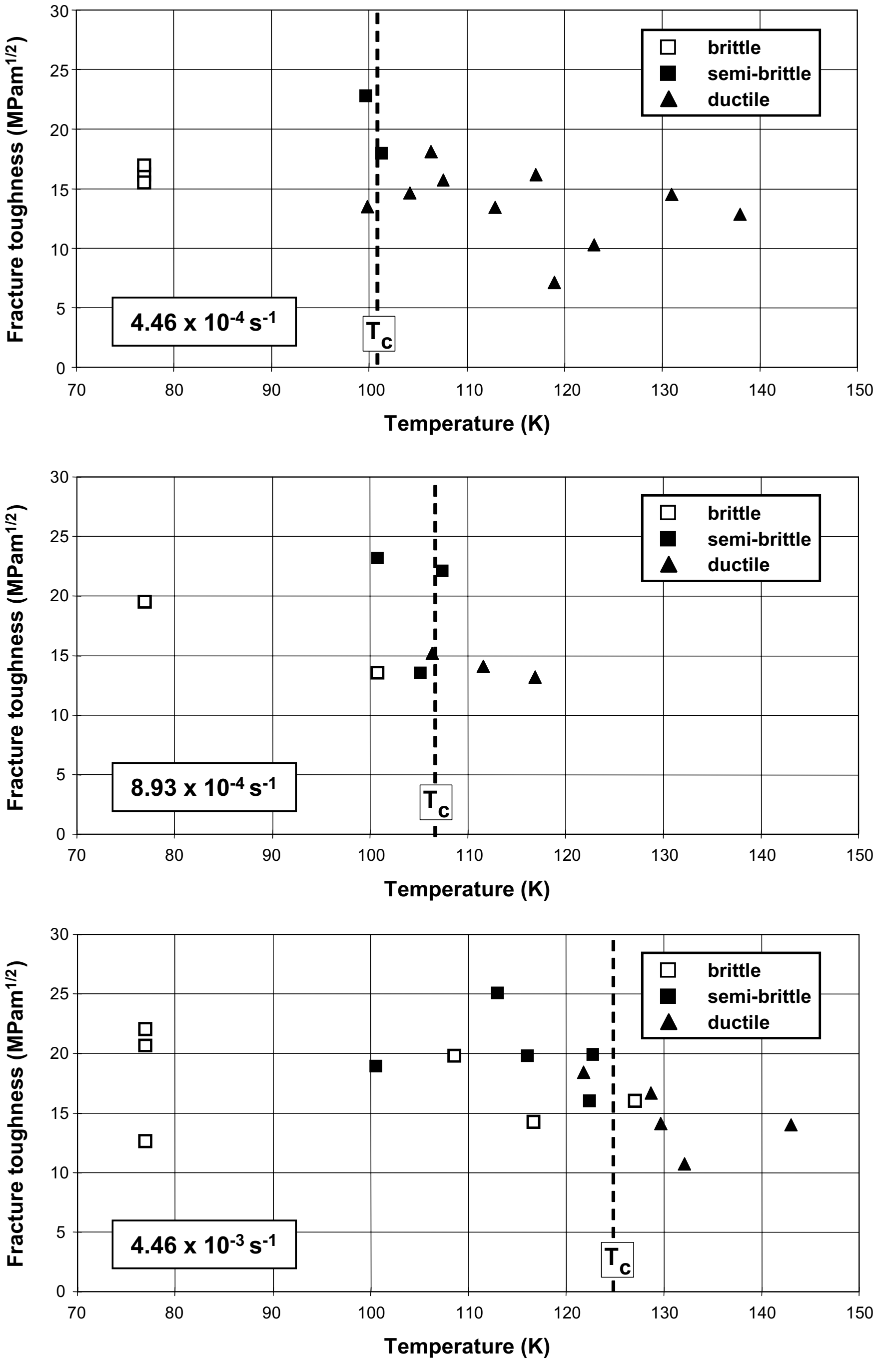

Fig. 3. Bend test results for polycrystalline iron–9% chromium at three strain rates.

at the highest strain rate used ($4.46 \times 10^{-3}\,s^{-1}$), at temperatures just above T_c. In one specimen (Fig. 4(a)), a twin has propagated from the notch in one grain, and has triggered twinning in a second grain; in another (Fig. 4(b)) slip lines can be seen beneath the notch root, and also emanating from the tip of one of cracks in the notch root, which has become blunted. Though the specimen exhibiting slip lines was tested at a slightly higher temperature (129 K) than the one exhibiting twinning

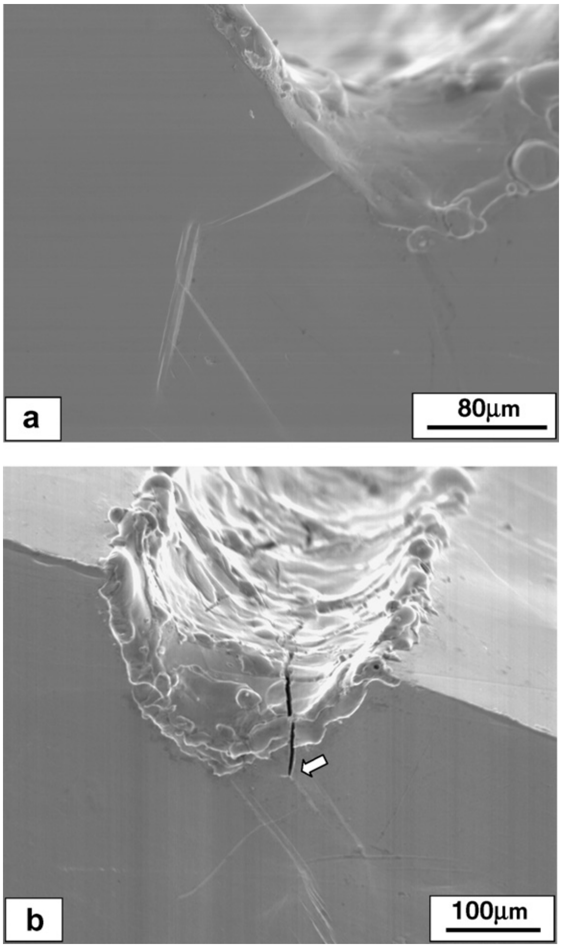

Fig. 4. Iron 9% chromium polycrystalline specimens tested at a strain rate of $4.46 \times 10^{-3}\ s^{-1}$: (a) 129 K (ductile) showing twins at the notch root; (b) 130 K (ductile) showing slip bands emanating from the notch root and from a crack tip (arrowed).

(128 K), the differences in behavior seen are most likely due to differences in orientation of the grains present in the observed region of the specimens.

For one test on an iron–9% chromium specimen, at 119 K and $4.46 \times 10^{-4}\ s^{-1}$, a load drop characteristic of semi-brittle behavior was noted at a tensile surface stress of 557 MPa ($\cong 8.77\ MPa\ m^{1/2}$ for short cracks in the notch root).The test was halted at a stress of 1250 MPa ($\cong 20\ MPa\ m^{1/2}$ for short cracks in the notch root), with the specimen remaining unfractured. SEM examination (Fig. 5(a)) showed the crack had propagated for several 100 μm below the notch base. After unloading, the specimen was re-tested at 77 K (Fig. 5(b)). Fracture occurred from the extended cracks at 1291 MPa, well above the low-temperature fracture stress and close to the maximum stress used at 119 K; that is,

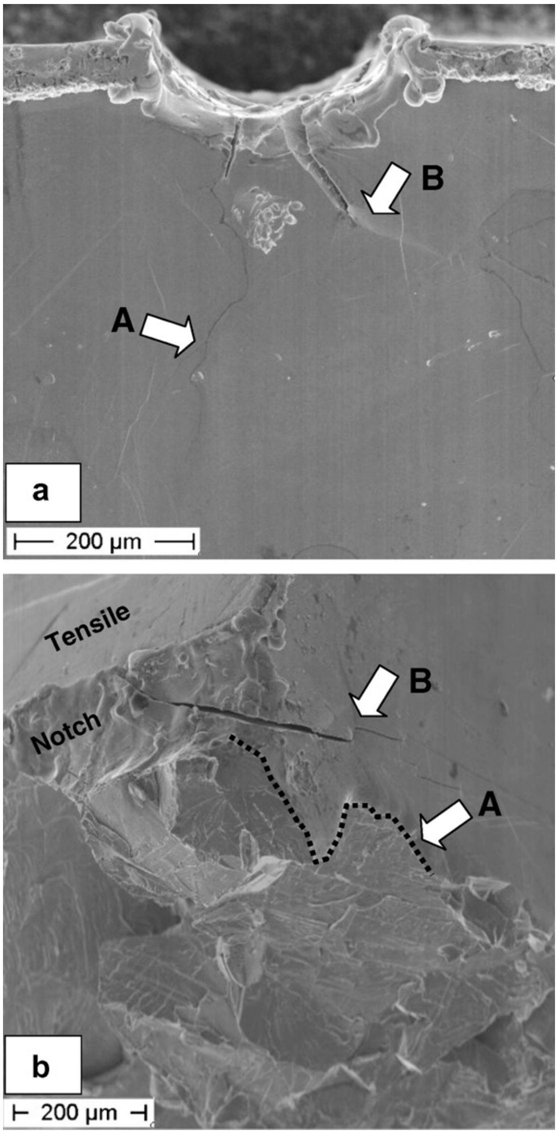

Fig. 5. Iron 9% chromium polycrystal, tested at 4.46×10 $10^{-4}\ s^{-1}$: (a) side face of specimen, after loading to 1250 MPa at 119 K. Cracks (A and B) extend from the notch base; (b) oblique view of fracture surface of the same specimen, after fracture at 77 K. Fracture has initiated from crack A.

crack shielding and/or blunting at 119 K has produced a ‘warm-pressing effect’ [25,26].

4. Summary

Experiments so far have shown that miniature specimens can give useful data about the fracture of bcc metals. The activation energy for the brittle–ductile transition in vanadium (~0.26 eV) appears to be lower than that for dislocation motion

(0.55 eV), in contrast to findings for many other materials. In polycrystalline iron the activation energy was found to be ∼0.21 eV, and in polycrystalline iron–9% chromium, ∼0.11 eV. The brittle–ductile transition behavior of single crystal vanadium is unusual for metals, showing a transfer from purely brittle to ductile behavior with no rise in fracture toughness except at temperatures very close to the transition. Iron and iron–9% chromium show semi-brittle behavior with elevated fracture toughness over a wider temperature range, but with no clear trend with temperature. Work continues on fracture and on dislocation behavior, so that the fracture behavior of these materials can be understood within the framework of existing dislocation-dynamic models. This is aimed at prediction of effects of radiation on fracture behavior, via its effects on dislocation mobility.

Acknowledgement

The work reported here was funded by UKAEA Culham; our thanks also to Professor S.L. Dudarev for useful discussions.

References

[1] A. Kohyama, A. Hishinuma, D.S. Gelles, R.L. Klueh, W. Deitz, K. Erlich, J. Nucl. Mater. 233–237 (1996) 138.
[2] K. Ehrlich, Fusion Eng. Des. 56–57 (2001) 71.
[3] H. Bolt, V. Barabash, G. Federici, J. Linke, A. Loarte, I. Roth, K. Sato, J. Nucl. Mater. 307–311 (2002) 43.
[4] B. van der Schaaf, D.S. Gelles, S. Jitsukawa, A. Kimura, R.L. Klueh, A. Möslang, G.R. Odette, J. Nucl. Mater. 283–287 (2002) 52.
[5] N.M. Ghoniem, S.H. Tong, B.N. Singh, L.Z. Sun, Phil. Mag. 81 (2001) 2743.
[6] B.N. Singh, N.M. Ghoniem, H. Trinkaus, J. Nucl. Mater. 307–311 (2002) 159.
[7] M. Ellis, The brittle–ductile transition in molybdenum, D.Phil thesis, University of Oxford, 1991.
[8] D. Hull, P. Beardmore, A.P. Valintine, Phil. Mag. 14 (1965) 1021.
[9] P. Gumbsch, J. Riedle, A. Hartmaier, H.F. Fischmeister, Science 282 (1998) 1293.
[10] A.S. Booth, S.G. Roberts, Acta Mater. 45 (1997) 1017.
[11] F.C. Serbena, The brittle–ductile transition in NiAl, D.Phil thesis, University of Oxford, 1995.
[12] P. Ochmann, H. Vehoff, P. Neumann, in: R.J. Arsenault, D. Cole, T. Gross, H. Sizek, P. Liaw, S. Parameswaran, G. Kostorz, Johannes Weertman Symposium, Minerals, Metals & Materials Soc., Warrendale, PA, USA,1996, p. 217.
[13] A.S. Booth, S.G. Roberts, J. Am. Ceram. Soc 77 (1994) 1457.
[14] F.C. Serbena, S.G. Roberts, Acta Metall. Mater. 42 (1994) 2505.
[15] R.O. Ritchie, J.F. Knott, J.R. Rice, J. Mech. Phys. Solids. 21 (1973) 395.
[16] C.St. John, Phil. Mag 32 (1975) 1193.
[17] S.G. Roberts, J. Samuels, Proc. R. Soc. Lond. A 421 (1989) 1.
[18] H.S. Kim, S.G. Roberts, J. Am. Ceram Soc 77 (1994) 3099.
[19] S.G. Roberts, in: H.O. Kirchner, L.P. Kubin, V. Pontikis (Eds.), Computer simulation in materials science – nano/meso/macroscopic space and time scales, NATO ASI series, series E (applied sciences), vol. 308, Kluwer Academic Publishers, Dordrecht, Netherlands, 1996, pp. 409–434.
[20] P.B. Hirsch, S.G. Roberts, J. Samuels, Proc. Roy. Soc. Lond. A 421 (1989) 25.
[21] M. Brede, Acta Metall. Mater. 41 (1993) 211.
[22] P.B. Hirsch, S.G. Roberts, Phil. Trans. Roy. Soc. 355 (1997) 1991.
[23] S.G. Roberts, S.J. Noronha, A.J. Wilkinson, P.B. Hirsch, Acta Mater. 50 (2002) 1229.
[24] C.T. Wang, D.W. Bainbridge, Met. Trans. 3 (1972) 3161.
[25] S. Tetelman, A.J. McEvily, Fracture of Structural Materials, John Wiley, New York, 1967, p. 338.
[26] A.S. Booth, M. Cosgrave, S.G. Roberts, Acta Metall. Mater. 39 (1991) 191.