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An experimental fracture map for uranium

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A two-dimensional (Ashby) *fracture mechanism map* with normalized stress versus homologous temperature has been constructed for metallic uranium from published experimental data. Uranium exhibits many modes of fracture so that the map is of particular interest. The map also has general relevance for non-cubic polycrystalline materials.

1. Introduction

The use of maps to describe the mechanical behaviour of materials developed by Ashby [1] is becoming quite widely accepted [2–4]. This general concept was first explored to map the *flow* behaviour of materials, and *deformation maps* were thus constructed. The deformation rate of a material is a function of shear stress, temperature, and material variables. Thus, in a space of stress and temperature, there will be a unique strain rate at each point. Furthermore, it is possible to calculate the strain rate due to each deformation mechanism; diffusion creep, dislocation creep, dislocation glide, twinning, etc. The mechanism map for deformation is constructed by finding the fastest deformation mechanism at each point on the map, and showing the areas where each mechanism thus dominates.

The extension of this concept to the *fracture* of materials has been examined recently [5]. A *fracture map* is not as easy to construct as a deformation map; fracture depends on more variables than flow. The *state* of stress, and loading history are, for example, of critical importance; this is immediately evident in considering the effect of cracks, and of cyclic loading. The effects of these limitations have been discussed in some detail elsewhere [5, 6]. However, there remains merit in considering a map based on the steady-stress, isothermal loading, in simple tension, of smooth bar specimens. In this case, there are still two variables, stress and temperature; and maps can be drawn in the space defined by these two variables. Both are non-dimensionalized, which makes for easier comparison between materials; stress is normalized against the elastic modulus, and temperature against the melting temperature. In making fracture maps, the dominant failure mechanism is determined, not from a comparison of constitutive equations, but from metallographic observation.

Uranium was chosen as a model material because its complex, orthorhombic, crystal structure, together with the two phase transformations, lead to a large number of different fracture modes. In fact, there has been little interest in the mechanical properties of metallic uranium since oxide and carbide reactor fuels became popular, but the material and the map has general relevance for non-cubic materials.

2. The fracture map

The experimental fracture map for uranium is shown in Fig. 1. Symbols on the map mark individual tests; open symbols are transgranular fractures, closed symbols intergranular. Each symbol has a number associated with it; these are the logarithm of the time to fracture in seconds. Let us now consider the individual fields within the map, remembering that the boundaries shown are valid only within the range of the data points. The curvature of boundaries in particular, is open to question, and this problem will be discussed later.

2.1. Twin parting/cleavage

At low temperatures, the complex crystal structure (orthorhombic) α-uranium deforms most

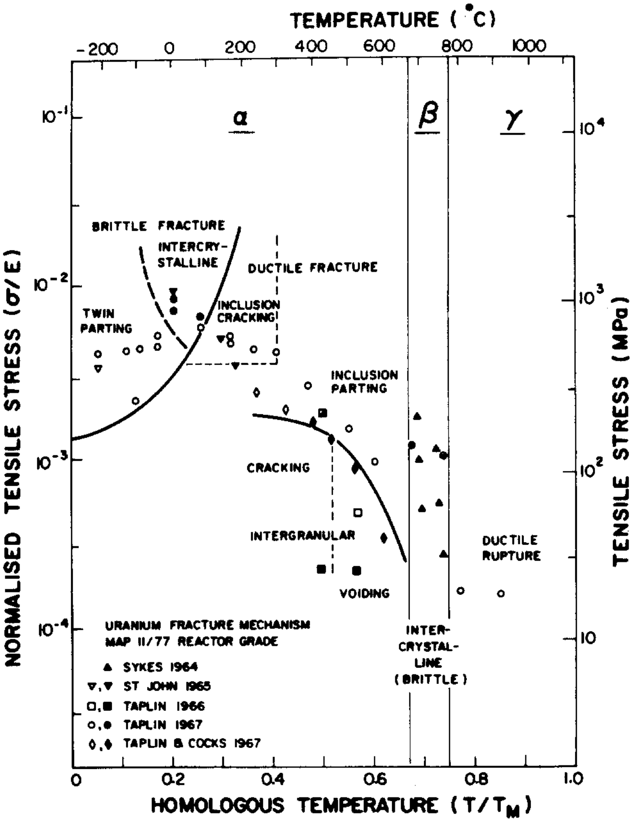

Figure 1 Fracture mechanism map for reactor-grade uranium. Numbers associated with experimental points are logarithm of the time to fracture in seconds. The boundaries are drawn on the basis of 50 μm grain size; points outside their fields are largely for 200 μm grain size.

easily by twinning. The growth of twins causes large stress concentrations at their tips, and the twin boundary provides a low energy path for brittle fracture. The stress required for large scale dislocation glide is up to an order of magnitude higher than the twinning stress [7], so that plastic blunting at the twin apices is resisted. Thus microcracks form along twin boundaries, as shown in Fig. 2, and fracture occurs with no significant ductility. The twinning stress increases with increasing temperature defining the right-hand boundary of the fracture mechanism field.

2.2. Ductile fracture

As the temperature increases, the stress required for dislocation glide decreases, until at $0.2\,T_\mathrm{m}$ (~300 K), it becomes less than the twinning stress. Large-scale plastic yielding leads to ductile failure with holes nucleating at large cuboid inclusions. These holes grow with plastic straining, until

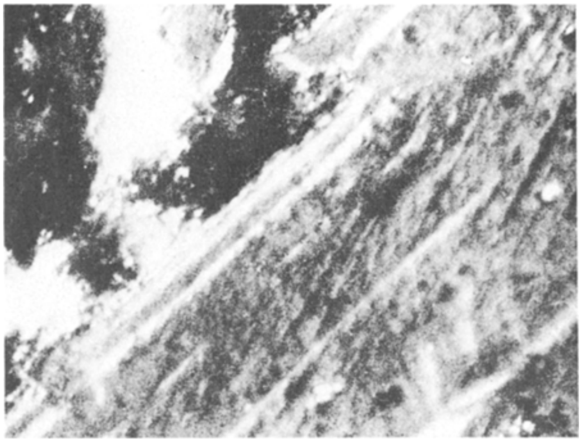

Figure 2 Twin-parting fracture at −95° C. This is a free fracture surface, × 800.

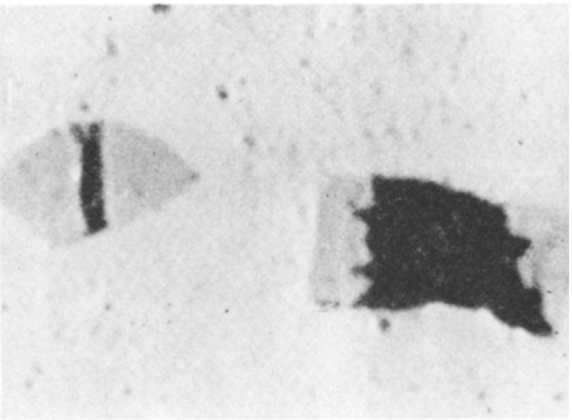

Figure 3 Cracking of cuboid inclusions at 50° C, × 2000.

catastrophic linkage occurs. There are two possible mechanisms for hole nucleation; the cuboids can crack internally, as shown in Fig. 3, or the inclusion–metal interface may decohere. More strain is required for decohesion, so that the tensile ductility is higher when internal cracking of inclusions is absent.

2.3. Low temperature intercrystalline cracking.

In the temperature region near $0.3\,T_\mathrm{m}$, intercrystalline cracks are sometimes observed, especially for coarse-grained materials; the data of St John [8] is for 200 μm grain size, while that of Taplin [9] is for 50 μm. In this temperature range, uranium is at its strongest, and the stress levels reached are in the same order as those predicted by the Smith–Barnby modification of Stroh's intercrystalline cracking model. A typical crack is shown in Fig. 4.

2.4. High temperature intergranular fracture

At temperatures above $0.4\,T_\mathrm{m}$, where grain boundary sliding can operate at a finite rate, fracture can occur by the formation of cracks and cavities on grain boundaries. In uranium, and for the short-term (~ 100 h) creep tests employed, the ductility is not greatly impaired by an intergranular fracture path. It is believed that all fractures in this region are initiated by the formation of very small cavities, which grow, by diffusion [10, 11] or viscous flow [12, 13] to form short cracks, of the order of a grain facet in length. At low temperatures, these cracks extend without blunting, maintaining a very large aspect ratio (length/height > 10). This mode is termed "brittle intergranular creep fracture", and is shown in Fig. 5. At higher temperatures the cracks blunt markedly and grow by plastic flow in the stress direction, to aspect ratios much less than 1. This

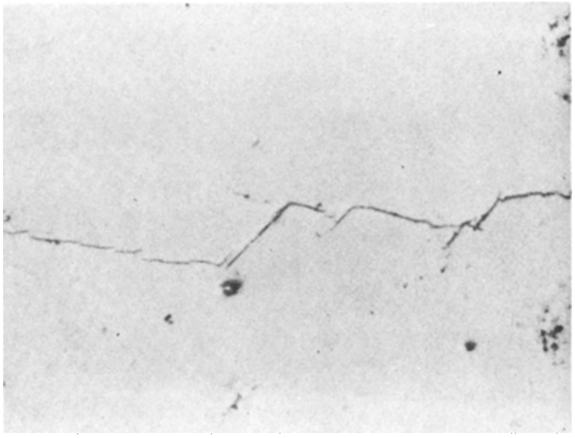

Figure 4 Intercrystalline crack at −10° C, propagating from the surface, × 100.

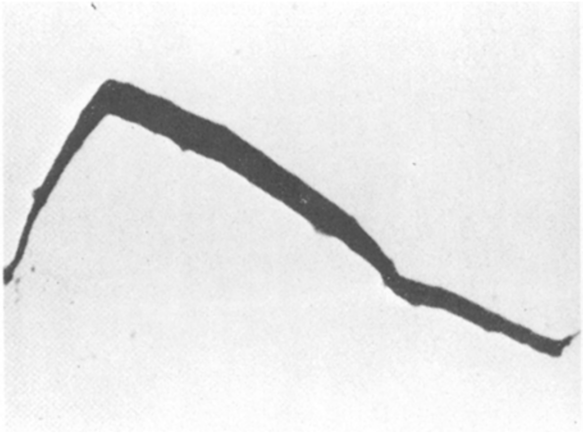

Figure 5 Intergranular crack formed at 420° C and a strain rate of 10^{-6} sec^{-1}, × 800.

is shown in Fig. 6 and is termed "ductile intergranular creep fracture". Fracture in this second case occurs by coalescence of a void sheet, as the legend on the map indicates.

2.5. Beta-uranium

At 933 K, uranium changes crystal structure to a yet more complicated lattice; a large tetragonal cell. This appears to have very little capacity to deform plastically, and fractures along grain boundaries. In this it is similar to certain ceramics. Sykes [14] has suggested that the high flow stress and low ductility of the beta-phase are related to its high stacking-fault energy. They might equally be related to the large burgers vector of dislocations (~ 10 μm).

2.6. Gamma-uranium

At 1048 K, a second phase change occurs, to very soft body-centred cubic, gamma-phase. No "fracture" process, as such, has been observed in this phase. Failure occurs by plastic instability, with a neck forming and thinning to a point to give "ductile rupture". The flow stress in this phase, at the very high temperatures required, is apparently too low to allow any significant cavity nucleation.

3. Discussion

The fracture mechanism map for alpha-uranium shows three broad fields; (a) brittle fracture at low temperatures; (b) transgranular ductile fracture at high strain rates and ambient to elevated temperature; (c) intergranular fracture at high temperatures and low stress. The brittle cracking fields are directly related to the formation of deformation twins, at a stress which increases with temperature. Near ambient tempeature, the intersection of a twin with a grain boundary nucleates an intercrystalline crack. The nucleation condition is given by Smith [14, 15] as:

$$\sigma^2 = \frac{4G\gamma_{\text{eff}}}{\pi(1-\nu)d} \tag{1}$$

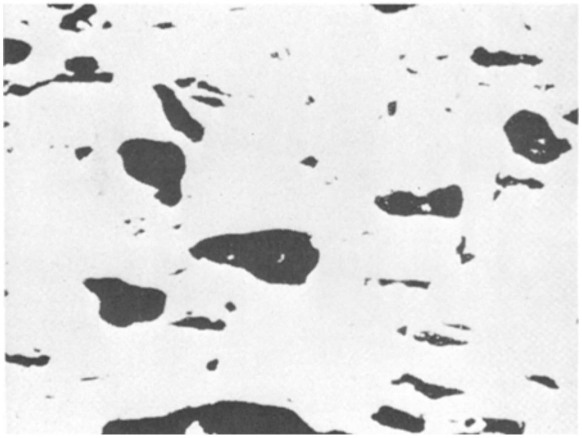

Figure 6 Intergranular voids formed in creep at 500° C and a strain rate of 10^{-5} sec^{-1}. The voids are strongly elongated in the direction of stress, × 470.

where σ is the nucleation stress for cracking, d is grain size, ν is Poisson's ratio, G is the shear modulus and γ_{eff} is the effective grain boundary fracture energy. Applying this relation to the data of Taplin and Martin [7], for 50 μm grain size, and that of St John [8] on material of 200 μm grain size, leads to an effective fracture energy of 100 Jm^{-2}. This is a nucleation condition only; Taplin and Martin show a long, non-fatal crack in 50 μm grain size material, 10 facets or so in length. This implies a fracture toughness for the material at this temperature of ~30 $\text{MPam}^{1/2}$ (30 Gr *), corresponding to a propagation energy of 6×10^3 Jm^{-2}. The reason for the nucleating energy being so low has been explained [9] in terms of large residual stresses arising from thermal cycling or phase transformation.

At lower temperatures the only cracks visible, apart from the single fracture surface are at the tips of stopped twins (Fig. 7). These cracks are of the order of 30 μm in length. The absence of longer cracks indicates that the twin cracks are only just sub-critical. If this is the case, the fracture toughness of the metal at 150 K is of the order of 5 Gr, giving a propagation energy of 150 Jm^{-2}. Thus, the reason for the transition from intercrystalline fracture to twin-parting cleavage is a reduction of the crack propagation energy for twin-parting below the nucleation energy for intercrystalline cracks. The propagation energy for twin-parting is of the same order as the cleavage energy in steels [17], indicating a contribution of plasticity to the fracture process. This is consistent with the observation of an opening ~2 μm of the stopped-twin cracks [18].

Figure 7 Crack at the end of a stopped twin, formed at −95° C. The crack is 30 μm long, with an opening of ~3 μm, × 850.

At higher temperatures, where dislocation glide takes over from twinning as the dominant deformation mechanism, the fracture process becomes ductile. In this regime, the strain to fracture has two components; the nucleation strain required to crack inclusions, or to separate them from the matrix; and the growth strain required to enlarge the resulting voids until they can coalesce by plastic collapse, and cause final fracture.

At temperatures less than 300° C, the nucleation strain is very small (<2%) [7], and void initiation occurs by cracking of large cuboid inclusions, U(O, C, N). Above 300° C, where thermally-activated flow starts, crack initiation is by the decohesion of the inclusions, requiring appreciable strain. This higher temperature mechanism is consistent with the recent work of Butcher [19], who examined the initiation of voids at hard particles in a plastic material. The initiation mechanism depended on inclusion aspect ratio, as it does here, and was essentially strain controlled, both in a model system and for MnS in steel. If the work of Butcher describes accurately the nucleation strain of ~20% for higher temperature fracture, another explanation must be found for the initiation mechanism. Since alpha-uranium is of orthorhombic crystal structure, the number of initial glide systems is limited; it is thus possible that dislocation pile-ups could occur at inclusions, without being released by secondary slip systems. Smith and Barnby [20] have formulated the condition for cracking of inclusions intersecting a slip band, as:

$$\sigma_c\sqrt{\pi\lambda/2} = K_c\ \text{(inclusion)} \qquad (2)$$

where 2λ is the interparticle spacing, σ_c is the crack nucleation stress and K_c is the fracture toughness of the inclusion.

For a 5% volume fraction of 10 μm inclusions, $2\lambda = 45$ μm. The critical stress, from the data of Cocks and Taplin [21] thus would be ~240 MPa. Thus the inclusion toughness is ~1 Gr, typical for a covalent crystal. This critical stress is always exceeded in tensile tests at temperatures below 300° C.

The growth strain depends on the volume fraction (v_f) of nucleating particles, which for uranium containing 1000 p.p.m. of C + N + O is

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*1 Gr (griffith) = 1 $\text{MPa m}^{1/2}$. This unit has been proposed in [16].

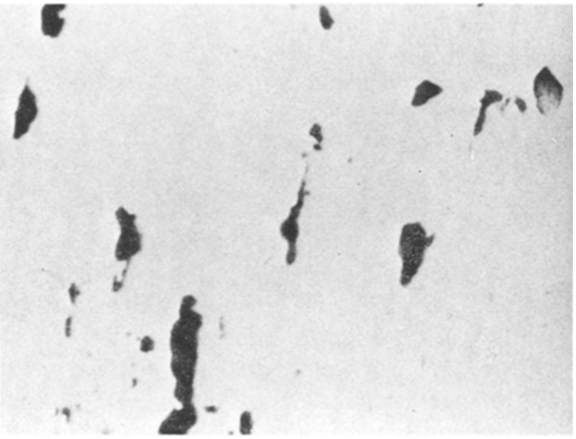

Figure 8 Transgranular voids formed in a slow tensile test at 500° C and a strain rate of 10^{-3} sec^{-1}. The voids are strongly elongated, as in Fig. 6, × 700.

approximately 4 to 5%. The most commonly used model is that of Brown and Embury [22], or Thomason [23], which assumes growth of the void proportional to the macroscopic strain, and a geometric coalescence criterion in which the void "height" equals the ligament width. The two models differ slightly in detail but predict that the growth strain (ϵ_g) is given by:

$$\epsilon_g = \frac{1}{c}\left(\frac{a}{V_f^{1/2}} - b\right). \qquad (3)$$

Brown and Embury have $a = 0.72$, $b = 0.82$, $c = 1.8$. and Ashby [24] has fitted experimental data to Equation 3 giving $a = 0.7$, $b = 1.0$, $c = 1.8$. The larger value of c implies some amplification of strain at the void, which is consistent with the experimental observations of hole growth by Finnie [25]. Substituting $V_f = 0.05$ and Ashby's constants, the predicted growth strain is 0.79. However, the experimental growth strain is only 0.3, which indicates that the growth strain for large voids is much less than for small voids; plastic collapse occurs long before the geometric condition is satisfied. The large ligaments (45 μm wide) fail by necking, rather than slip band formation; thus voids "see" each other earlier when they start large. The failure is then analogous to a number of discrete tensile specimens pulled in parallel.

At higher temperatures the opposite problem is found; the growth strain is *larger* than the Ashby predictions, and the ligament shape remains stable even when the ligament height is much greater than its width (Fig. 8). Reduction of area up to 85% is seen, corresponding to a fracture strain of 1.9, and the volume fraction of voids at fracture is 20%. Similar observations have been seen in aluminium alloys [26] and copper alloys [27]. The stability of the ligaments is attributed to local strain-rate sensitivity in the cubic metals, and a similar explanation is presumably applicable here. There is little evidence of macroscopic strain rate sensitivity since the uniform strain decreases as the fracture strain increases with increasing temperature.

This high ductility persists into the intergranular fracture field. These are two distinct subfields, defined basically by the 450° C isotherm. Below this temperature intergranular cracks remain sharp, growing along grain boundaries normal to the applied stress. At higher temperatures they become extremely elongated parallel to the stress direction, and like transgranular voids, fail to coalesce until the ligament height is much greater than the width. The boundary between transgranular and intergranular fracture fields show a break at this temperature, which is only slightly higher than the start of the strong temperature dependence of tensile strength and the likely onset of marked strain-rate sensitivity.

The "cracking" mechanism is remarkably independent of stress; it operates at least as well at 28 MPa as at 300 MPa. This is quite unlike the behaviour in f c c metals [28] where the *cracking* mechanism tends to operate at high stress, and *discrete cavities* tend to form at low stress. This observation, and the enormously elongated voids at higher temperature, suggests a change in deformation mechanism, at least on a microscopic scale, with change of temperature. Sharp cracks normally imply a limitation of "plastic" relaxations of local stress concentrations, and minimal strain-rate sensitivity. However, a model

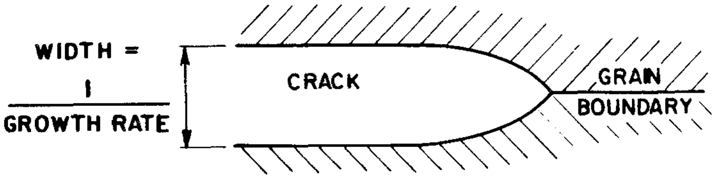

Figure 9 The shape of an intergranular crack, growing by vacancy condensation at a rate controlled by surface diffusion, as modelled by Chuang and Rice [29].

has been developed [29] for the growth of a crack by vacancy diffusion, which predicts a crack with parallel faces, with a curved "cusp" at the tip (Fig. 9) remarkably similar to the observed crack profile (Fig. 5). Similarly, elongated voids imply very high strain-rate sensitivity, which is satisfied better, near a void, by dislocation motion than by diffusion creep. Vacancies tend to condense on the growing void, modifying its shape away from the elongated "viscous" void. Dislocations, on the other hand, in the absence of sub-cell formation, can allow high strain-rate sensitivity [30]. It is proposed therefore that the transition in intergranular fracture mechanisms is caused by a transition in microdeformation mechanism from diffusion (Coble) creep to dislocation creep. This alone makes it clear that a deformation mechanism map is, at least, a very useful adjunct to a fracture mechanism map. It does not seem possible to construct such a deformation map for uranium with the available (published) data. It is very sparse, and to construct a mechanism map would require assumptions about the mechanisms themselves which could not be justified for a metal of unusual crystal structure. Deformation mechanisms must be deduced from the mechanical behaviour, whereas fracture mechanisms can be discerned directly.

The fracture of the beta-phase is essentially brittle, but there is a finite and measurable fracture strain. The intercrystalline fracture, appreciable variation of fracture stress with strain rate, low ductility and high creep strength are reminiscent of ceramics at similar homologous temperature. This is not surprising, since bonding in uranium has considerable covalent character, and the crystal structure of the beta-phase is very complex. Thus, deformation is difficult, and does not easily accommodate incompatibilities at grain boundaries, leading to fracture at small strain. The gamma-phase is body-centred cubic, and deforms at stresses so low that no fracture nuclei form, and fracture is essentially by plastic collapse, or rupture, with very large strains, ~5, in the neck. This is common in cubic metals at high temperatures, unless void nuclei pre-exist.

Comparison of the uranium map with a map for an fcc or bcc material, shown schematically in Fig. 10, (after Ashby [24]) shows that the general shape of the map is similar to the cubic metals. The differences, primarily in the shape of the intergranular fracture field and the position of the brittle field, are due to the crystal structure itself, as are the multiple phase fields. There appears to be some utility in constructing fracture maps for the less-common crystallographic systems. Hexagonal metals are obviously of practical interest; magnesium, zinc and zirconium have considerable application. Ceramics are of course of major interest now and since the fracture properties of most ceramics are less well known, the construction of approximate fracture maps would seem to be a good starting point for the characterization of their behaviour.

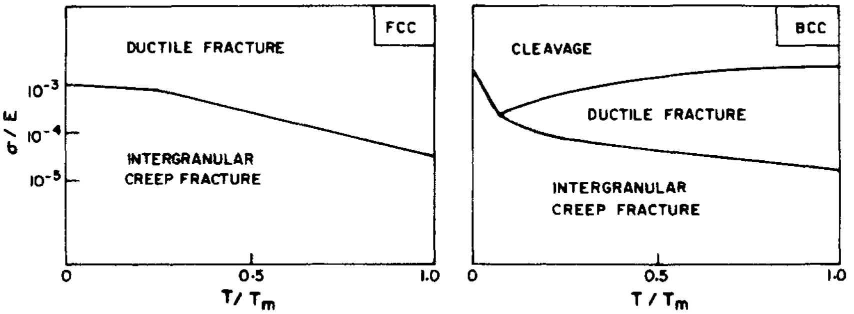

Figure 10 Schematic fracture mechanism maps for: (a) face centred cubic; (b) body centred cubic, after Ashby [1].

Acknowledgements

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