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TOUGHNESS VARIATIONS DURING THE TEMPERING OF A PLAIN CARBON, MARTENSITIC STEEL

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INTRODUCTION

The aim of this paper is to present results of toughness measurements and fractographic observations made on a quenched and tempered 0.6%, carbon steel and to use these as a basis for a discussion of the factors affecting "623K (350°C) embrittlement" (one-step temper embrittlement). When a martensitic steel is tempered, it is generally observed that its toughness, as measured by room-temperature Charpy values, drops noticeably for tempering temperatures in the range 550-675K. Work by Lement et al [1] associated the effect with the formation of cementite at martensite packet boundaries, suggesting that the fracture would be intergranular with respect to packet boundaries, but transgranular with respect to austenite grains, except where packet and grain boundary coincided. Klingler et al [2] suggested that the formation of cementite at prior austenite boundaries led to the production of ferritic grain-boundary networks by carbon denudation, so promoting fractures which were intergranular with respect to the prior austenite grains. In discussion [3] Lement agreed with the intergranular nature of the fracture, but ascribed it to the cementite, rather than the ferrite network. Neither set of authors presents any fractography and their toughness minima are taken from room-temperature Charpy values. It is interesting to note that the results for Klingler's 4340 steel (OQ 1113K) barely show a minimum throughout the tempering range 420-675K and that at no stage in the range does the toughness drop below 22J. On comparing these values with full transition curves, it can be seen that they are not much less than the *upper shelf* levels and that any minimum is therefore associated with a fracture which is substantially fibrous in character. The improvement in toughness at higher tempering temperatures is attributed to general matrix softening.

A variation on the cementite growth mechanism as a cause of 623K embrittlement is suggested by McMahon in his plenary paper [4]: the coarsening of M_3C rejects impurity element at prior austenite grain boundaries, which enhances any tendency for intergranular fracture. The results for commercial 4340 steels (OQ 1123K) show minima at about 625K, but the minimum is absent for a pure steel, whose toughness is about 25J in the range 420-625K. No fractography is included but the impure steels presumably give intergranular fractures at approximately 625K, compared with a mixed fibrous/cleavage fracture in the pure steel. Impurity rejection giving rise to intergranular fracture in alloy steel specimens tempered in the range 550-675K is supported metallographically by Tait and Knott [5] and is suggested by Kula and Anctil [6] to explain their test results: see below.

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Further evidence on the cause of 623K embrittlement is confusing. McMahon and Thomas [7] attribute a minimum in K_{IC} values, during the tempering of a high-Cr steel, to the formation of coarse *inter-lath* cementite, but the minimum itself is not unambiguous and they present no fractography. Lindborg and Averbach [8] show, however, that the fracture path in thin films of a pure, tempered, Fe-0.3%C steel cuts *across* martensite laths rather than follow the boundaries, where coarse carbides are present.

Kula and Anctil [6] have compared a number of K_{IC} results for 4340 steel. At room temperature, *no minimum could be found*, although it was present when samples were tested at 200K. The room temperature toughnesses for 478K and 644K tempers were found to decrease markedly with total (S+P) content. No detailed fractography was reported. Similarly, Walker and May [9] could not find a minimum in room temperature K_{IC} values for different tempers in a NiCrMoV steel: "quasi-cleavage" is reported for a 473K temper and "mixed ductile-brittle manner" for 573K and 673K tempers. Horn and Ritchie [10] obtain a small drop in K_{IC} for a 4340 steel tempered at 573K, associated with a change from fibrous to intergranular fracture, and for 300M steel tempered at 673K, associated with a mixed fibrous/cleavage/intergranular appearance. Ritchie [11] has also observed a minimum for En30B (0.4%C 4.25%Ni), associated with intergranular fracture. Finally, Backfisch and Schwalbe [12] find a minimum, when the fracture appearance remains fibrous throughout.

From this survey of previous observations, it was felt that a clear example of the simple carbide coarsening mechanism had not yet been shown and that it was of interest to establish whether this basic mechanism could operate. The work described in the present paper therefore tries to answer three questions:

i) is it possible to obtain a "623K embrittlement" minimum in the graph of (valid) K_{IC} vs. tempering temperature, when the fracture mode remains intergranular cleavage throughout?

ii) does this transgranular cleavage cross martensitic laths or does it follow lath boundaries?

iii) is the improvement in toughness at higher tempering temperatures due simply to matrix softening?

EXPERIMENTAL

Preliminary results on a quenched and tempered 0.4%C steel, tested at 173K, showed a minimum in K_{IC} for tempering temperatures around 625K, but the steel had not transformed completely to martensite, and it was considered that the presence of free bainite or ferrite might confuse the interpretation of the results. Consequently, a composition of Fe-0.6%C was chosen, to try to ensure complete transformation to martensite on quenching, yet to minimize the amount of retained austenite in the microstructure. The chemical analysis of the steel was 0.57%C, 0.60%Mn, 0.18%Si, 0.022%S, and 0.023%P. Fracture toughness testpieces were machined as standard SEN bend specimens, having B = 18mm; see [13]. All specimens were austenitized for 1 h at 1223K and were tempered, in pairs, at various temperatures in the range 483-833K. They were then fatigue pre-cracked, following the standard procedure [13]. Fracture toughness tests were carried out at 273K and 173K, in low-temperature baths containing either ice/water or IMS/acetone cooled with liquid nitrogen, using a Mand servo-controlled electro-hydraulic testing machine. The temperature control at the lower temperature was ±2K. To obtain yield stress values, tensile specimens,

which had been heat-treated alongside the toughness testpieces, were tested at 273K and 173K, using a low-temperature rig attached to a Mand screw-driven tensile testing machine. Fracture surfaces were examined in a Cambridge Instruments scanning electron microscope, operating at a voltage of 30kV. Selected specimens were nickel-plated, to preserve the fracture edge, and were examined metallographically, using a Zeiss optical microscope and the scanning microscope.

RESULTS

The as-quenched microstructure was found to be almost completely martensitic, although, in some small, isolated areas, traces of higher temperature transformation products were visible at prior austenite grain boundaries.

The uniaxial yield stresses at 273K and 173K are shown as functions of tempering temperature in Figure 1. The yield stress levels at 273K, for tempering temperatures in the range 525-600K, are similar to ambient values for 4340 [6, 14] and other low-alloy steels, containing approximately 0.35-0.40%C [9, 15], but the lack of Cr, Mo, V or Si in the composition causes the steel to soften more rapidly at higher tempering temperatures. Low-temperature yield stress values for the alloy steels are sparse, but the present results at 173K do seem to be comparable with figures in the literature [6, 14, 15] even for tempering temperatures as high as 675K. The effect of lowering the testing temperature by 100K is to increase the yield stress more or less uniformly by about 170 MPa; alternatively, it may be seen that the 273K yield stress of a specimen tempered at 570K is about equal to the 173K yield stress of a specimen tempered at 655K (approximately 1600 MPa).

The fracture toughness values at 273K and 173K are plotted as functions of testing temperature in Figures 2 and 3 respectively. All results were valid K_{IC} figures, using the criteria laid down in [13]. There is a minimum in the 273K curve (Figure 2) corresponding to a tempering temperature of about 570K, and a slightly less clear minimum in the 173K curve (Figure 3) at about 665K.

Fractographic examination of the fracture surfaces, using the SEM, showed that *all* fractures, at 273K and 173K were *transgranular cleavage*, e.g. Figure 4. For tempering temperatures less than 770K, the whole surface showed flat, transgranular facets, but for higher temperatures, the surfaces were rougher and narrow (approximately 5% of the specimen thickness) shear lips were present.

Close metallographic examination was made of nickel-plated sections through the fractures which corresponded to the toughness minima at 273K and 173K (the 570K and 665K tempers). The transgranular cleavage nature of the fracture was confirmed and the fracture path appeared to run across martensite packets. This was shown more clearly by sub-surface micro-cracks, and Figures 5 and 6 are typical of the general observations. There was no tendency for the cracks to run around prior austenite grain boundaries, around martensite packet boundaries or along lath edges, except in a few, isolated cases where small portions of such paths were particularly favourably oriented with respect to the rest of the crack plane. Observations in the scanning microscope confirmed that the fractures ran across the laths, Figure 7.

DISCUSSION

The strength levels and microstructures obtained in this 0.6%C steel over the range of tempering temperatures associated with the toughness minima (570-665K) are comparable with those in 4340 and similar low-alloy steels, so that the system may be regarded as a simple model of general tempering behaviour, but devoid of alloying element - impurity element interactions.

With respect to the three questions posed in the introduction, the following conclusions may be drawn:

a) there do appear to be minima in the curves of K_{IC} vs. tempering temperature, at approximately 570K for 273K tests and approximately 665K for 173K, for fractures which are 100% transgranular cleavage. The minimum toughness obtained is about 50 MPa·m$^{1/2}$ in both cases. This compares with Kula and Anctil's [6] values for 4340 of approximately 45 MPa·m$^{1/2}$ at 200K and approximately 60 MPa·m$^{1/2}$ at room temperature, the latter in a range (dependent on purity) of 68 to 28 MPa·m$^{1/2}$. Hays and Wessel [14] obtained a value of approximately 50 MPa·m$^{1/2}$ at 210K for 4340 tempered at 480K. Horn and Ritchie [10] obtain a minimum room temperature value, at 720K, in air-cooled 300M (high Si) steel, of 45 MPa·m$^{1/2}$, associated with cleavage fracture.

b) the fracture path is transgranular with respect to the austenite grains and the martensite packets. This supports the results of Lindborg and Averbach [8], and is similar to observations made in lower carbon martensitic microstructures [16]. In this model system, the coarsening of inter-lath cementite therefore tends to enable larger crack nuclei to be formed, rather than providing a more favourable path for propagation. If impurity were rejected simultaneously, the propagation path would follow grain or packet boundaries, and the work of fracture would decrease. The low toughness values quoted by Kula and Anctil [6] (down to 28 MPa·m$^{1/2}$) presumably arise from this effect. It is tempting to assume that the cracks follow the path that they do because this represents the (001) cleavage plane in the martensite laths. An approximate crystallographic trace analysis of the cracks and laths in Figure 6 shows that this view is not inconsistent, if a $(225)_\gamma$ habit is assumed for the laths, but other possibilities may exist: for example, the path may follow the epsilon carbide habit plane within the laths; see also [8].

c) the improvement in toughness does seem to be attributable simply to matrix softening. Bearing in mind the errors involved in testing temperature (±2K), tempering temperature (±5K) and precise location of the toughness minimum, it seems that the shift in position of the minimum, from approximately 570K at 273K, to approximately 665K at 173K, occurs because the yield stress at 173K for a 665K temper is as high as that at 273K for a 570K temper (Figure 1). It might be thought that the value of K_{IC} at the 173K minimum should be less than that at the 273K minimum, because the 665K tempered microstructure would contain coarser carbides (larger crack nuclei) than the 570K microstructure. This argument, however, does not take into account the bimodal distribution of carbides: relatively coarse *inter-lath* cementite (first forming at approximately 525K [1, 2] and coarsening or increasing in volume fraction over the next 50K or so) and fine *intra-lath* epsilon carbide which transforms to cementite and coarsens with time, but, over the range of interest, is always very much finer than the inter-lath cementite. It is plausible that, over the range of 570K-665K, the intra-lath precipitate coarsens and gives rise to softening, but the inter-lath cementite does not coarsen significantly, particularly if its carbon content has been drawn primarily from regions of super-saturated, inter-lath, retained austenite [7] rather than from the regions of as-quenched martensite. Thus, the intra-lath distribution

governs the strength level: the inter-lath carbide size governs the size of any crack nucleus, so that the toughnesses of the 570K and 665K microstructures, *at constant yield stress*, should be similar.

The stress required to propagate a cleavage fracture in low-strength steels may be related to the microstructure in a fairly straight-forward manner, considering, for example, a cracked grain-boundary carbide as a Griffith crack, which spreads under the influence of the applied tensile (fracture) stress and stress fields from local dislocation arrays, see e.g. [17]. Curry [18] has recently examined the fracture of *well-tempered*, spheroidal carbide distributions, neglecting the dislocation contribution, and treating the problem as one of the Griffith-type propagation of an isolated penny-shaped crack of radius equal to that of the carbide particle. Hodgson and Tetelman [19] have used a similar approach, using a plane strain analysis, but make the point that the fracture stresses of microstructures tempered at the lower tempering temperatures are affected strongly by the distribution of fine intra-lath carbide particles.

The maximum width of the inter-lath cementite in the present steel at the temperature of the toughness minimum is likely to be of order 0.2-0.4μm [1, 2, 19], and of platelet form. Treating the propagation of a cracked nucleus as that of a Griffith crack, we obtain:

$$\sigma_F = \left[\frac{2E\gamma_p}{\pi(1-\nu^2)a}\right]^{1/2} \qquad (1)$$

where a is 0.1μm (0.2μm), E is 200 GPa, ν is 0.3, and γ_p, the "work to fracture" may be taken as 14 Jm^{-2} [17]. The resultant value of σ_F is then 4425 MPa (3130 MPa), which is a factor of 2 (1.5) higher than values for other steels [17]. From this value of σ_F and the values of K_{IC} (50 MPa·m$^{1/2}$) and σ_Y (1600 MPa) at the toughness minimum, it is possible to calculate values of the "critical distance", X_c, ahead of a crack, over which high stresses must be attained. Following [20], we find that $X_c/(K_{IC}/\sigma_Y)^2 \sim$ 0.015 (0.055), $X_c \sim$ 15μm (54μm). These figures are speculative, but are consistent with the view that high stresses must be attained over a distance equal to one or more packet widths, presumably in order to find a sufficiently thick carbide to act as a crack nucleus (cf. the packet width in Figure 6). The carbides are likely to be thickest at the high angle packet boundary, which is also perhaps where most retained austenite is to be found.

There are clearly further points of detail to be explored, but the present work has shown the existence of a toughness minimum during the tempering of a plain carbon steel, for fractures which are transgranular cleavage in all cases. If the coarsening of inter-lath cementite is regarded as the basic cause of 623K (350°C) embrittlement, the incidence of grain boundary fracture must be regarded as incidental, arising most probably from the rejection of trace impurity elements, during carbide growth and possibly from alloying element: impurity element interactions.

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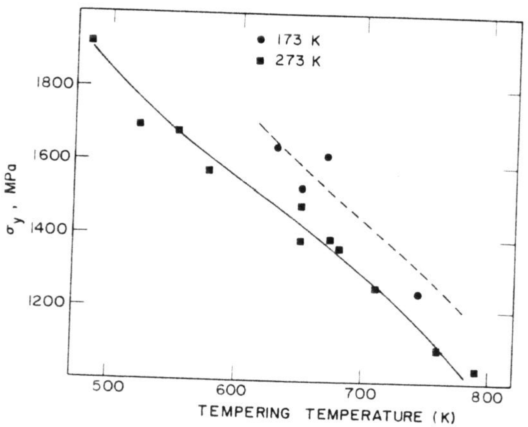

Figure 1 Variation of Uniaxial Yield Stress, at 273 K and 173 K, with Tempering Temperature

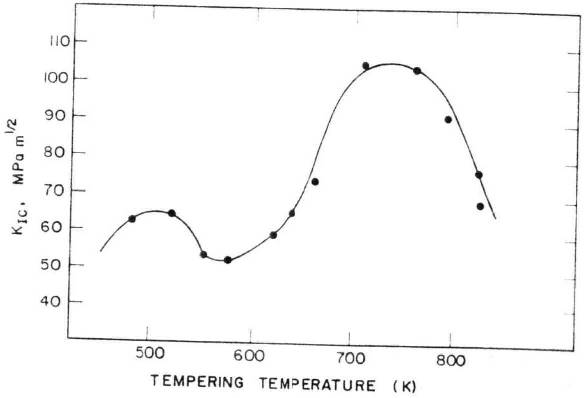

Figure 2 Variation of K_{IC}, Measured at 273 K, with Tempering Temperature

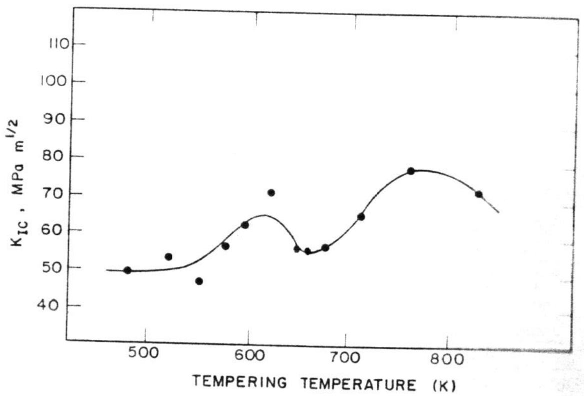

Figure 3 Variation of K_{IC}, Measured at 173 K, with Tempering Temperature

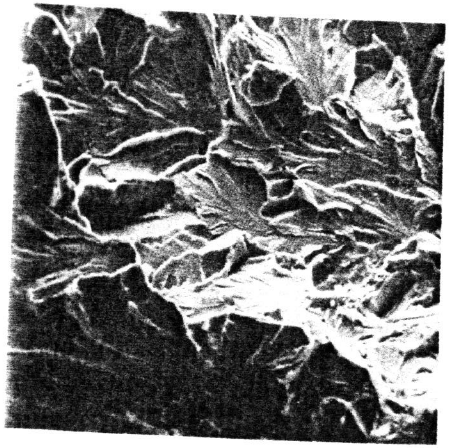

Figure 4 Scanning Electron Micrograph of Fracture Surface of Specimen Tempered at 833 K and Tested at 273 K. Magnification X 675

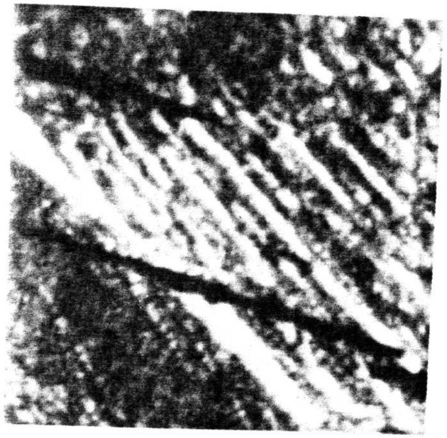

Figure 5 Sub-Surface Microcracks in Specimen Tempered at 683 K and Tested at 173 K. Magnification X 2860

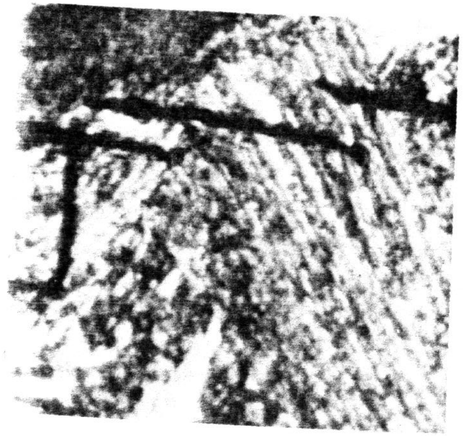

Figure 6 Array of Microcracks in Specimen Tempered at 683 K and Tested at 173 K. Magnification X 2860

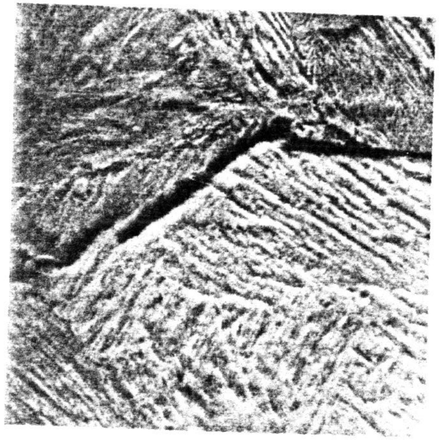

Figure 7 Scanning Electron Micrograph of Sub-Surface Microcracks in Specimen Tempered at 683 K and Tested at 173 K. Magnification X 1830