The Influence of Grain Size on the Mechanical Properties of Steel

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Abstract

Many of the important mechanical properties of steel, including yield strength and hardness, the ductile-brittle transition temperature and susceptibility to environmental embrittlement can be improved by refining the grain size. The improvement can often be quantified in a constitutive relation that is an appropriate variant on the familiar Hall-Petch relation: the quantitative improvement in properties varies with $d^{-1/2}$, where d is the grain size. Nonetheless, there is considerable uncertainty regarding the detailed mechanism of the grain size effect, and appropriate definition of "grain size". Each particular mechanism of strengthening and fracture suggests its own appropriate definition of the "effective grain size", and how it may be best controlled.

1. Introduction

The influence of grain size on the mechanical properties of steel is most commonly expressed in a series of constitutive equations that have the Hall-Petch form. Over the range of conventional grain sizes, the values of typical mechanical properties increase with the reciprocal root of the grain size. The classic Hall-Petch equation relates the yield strength to the grain size:

$$\sigma_y = \sigma_0 + K_y d^{-1/2} \tag{1}$$

where K_y is the Hall-Petch slope and d is the mean grain size. An identical relation holds for the indentation hardness. A similar equation applies to the cleavage fracture stress (σ_f) of high-strength steels,

$$\sigma_f = K_f d^{-1/2} \tag{2}$$

and an equation of the Hall-Petch form is also often useful for predicting the ductile-brittle transition temperature:

$$T_B = T_0 - K_B d^{-1/2} \tag{3}$$

The fatigue strength is often taken to vary with grain size in the same way.

The wide applicability of the generic Hall-Petch relation makes it one of the most important constitutive relations in Materials Science, and certainly the most common in mechanical metallurgy. However, its mechanistic origins and even its precise meaning remain imperfectly understood. For example, four distinct models have been advanced to justify eq. (1), each of which has at least a couple of variations. Three different models have been advanced to explain deviations from Hall-Petch behavior at very small grain sizes. The grain-size dependence of the cleavage stress is a straightforward consequence of both the Griffith and Orowan fracture criteria, but the Hall-Petch relation for the ductile-brittle transition presumes a linear connection between the fracture stress and T_B that is more difficult to justify.

Even given theoretical models that generate the Hall-Petch relation, there are ambiguities in its content. Particularly in martensitic steels, the meaning of the grain size is unclear. In fact, different measures of the grain size govern different processes; in some cases grain refinement techniques that produce dramatic improvements in cleavage resistance have little or no effect on strength and may actually reduce toughness after environmental embrittlement. The use of the *mean* as the measure of grain size is also difficult to justify, since yield and fracture ordinarily reflect the behavior of "weakest links" in the microstructure that should be associated with extremes in the distribution of grain size.

In the following we discuss the models that predict Hall- Petch behavior and the ambiguities that influence the choice of grain size, with emphasis on the strength and toughness of steel.

2. The Influence of Grain Size on Strength

There are at least four conceptually different models that lead to a Hall-Petch relation for the yield strength.

(1) The first, and most commonly cited, is the dislocation pile-up model [1]. Let an array of identical dislocations pile up against a grain boundary under the effective shear stress, $\tau_e = (\tau - \tau_i)$, where τ is the applied stress and τ_i is a local correction due to friction and back stresses. It can be easily shown that the stress at the head of the pile-up is

$$\tau_p = n\tau_e \tag{4}$$

If the pile-up has length, L, the number of dislocations it contains is also determined by the effective stress, and is

$$n = \frac{q\pi L\tau_e}{Gb} \tag{5}$$

where G is the shear modulus and q is a geometric factor of order unity. The length, L, cannot be larger than the grain size, d, and is usually taken to be d/2. Yielding is assumed to occur when the stress at the tip of the pile-up, τ_p, reaches the critical value necessary to nucleate slip across the grain boundary. At this point,

$$\tau = \tau_y = \tau_i + \left[\frac{2Gb\tau_c}{q\pi}\right]^{1/2} d^{-1/2} \tag{6}$$

Eq. (6) takes the form (1) when τ_y is multiplied by the Taylor factor, m, to convert it to the tensile yield strength, σ_y.

(2) While the pile-up model is superficially plausible, it suffers from the disadvantage that well-defined pile-ups are rarely observed, particularly in steels. It is, therefore, important to note that the Hall-Petch relation also follows from a more general model, based simply on the stress developed at the tip of a slip plane across the grain [2]. A slipped plane acts very much like a shear crack, which produces a crack-tip stress field of the form

$$\tau\,(r) = (\tau - \tau_i)q(d/r)^{1/2} \tag{7}$$

where r is the distance form the crack tip and q is a factor of order 1 that depends on the details of slip. Assuming yielding when the stress reaches a critical value, τ_c, at some distance, r_c, ahead of the boundary, we have the Hall-Petch equation in the form

$$\tau_y = \tau_i + (\tau_c r_c)d^{-1/2} \tag{8}$$

(3) A third interpretation is based on the correlation between grain size and dislocation density at yield. In the simple hardening models, the critical resolved shear stress to move a dislocation through a distribution of dislocations varies with the dislocation density, ρ, as

$$\tau_c = \tau_i + \alpha Gb\sqrt{\rho} \tag{9}$$

where α is a factor in the range 0.1-0.3 [3]. An increasing body of evidence suggests that this *basic hardening law* applies widely to the deformation of metals, including steels [3,,4], beginning from the yield strength. If this is true, and the Hall-Petch relation also holds at yield, the two must imply one another, that is, the dislocation density at yield (ρ_y) must be determined by the reciprocal grain size according to the relation

$$\rho_y = \left[\frac{k_y}{\alpha G b}\right]^2 \left[\frac{1}{d}\right] \tag{10}$$

Two different models have been suggested to justify behavior like that in eq. (10). The first is an older model due to Li [5] who suggested that the dislocation density within the grain might be determined by sources located in the boundary. If these are distributed with a constant mean areal density, the number of dislocations within the grain (N) would be proportional to the grain boundary area, cd^2, where c is a constant that depends on the shape of the grain. It follows that the dislocation density is

$$\rho = \frac{N}{V} = \frac{cd^2}{d^3} = \frac{c}{d} \tag{11}$$

More recent interpretations [6] have been based on the distribution of "geometrically necessary" dislocations [7] that are required to assure compatible strain across grain boundaries. The number of geometrically necessary dislocations needed to accommodate a given misorientation across a boundary is proportional to the area of the boundary. Hence the expected number of geometrically necessary dislocations within a grain of size, d, after a given strain should be proportional to the grain boundary area, as in eq. (11), again giving a dislocation density that increases with the reciprocal grain size.

(4) A fourth, and unexpected source of Hall-Petch behavior is found in the statistics of the microstructural resistance to dislocation glide. The common bulk hardening mechanisms (solute hardening, forest dislocation hardening, precipitation hardening) can often be modeled by a process in which dislocations glide through a field of randomly distributed obstacles. The basic hardening law is an example; the dislocation density defines the obstacles to glide. In a real case a dislocation gliding through a grain has a finite length that is of the order of the grain size. Both theoretical analysis and computer simulation suggest that the stress required to force a dislocation across a plane of width, d, that contains a distribution of barrier obstacles increases as $d^{-1/2}$, just as in the Hall-Petch relation. The reason, in this case, is statistical. As the width of the array decreases it becomes easier to find configurations of obstacles that are anomalously strong in their resistance. Computer simulation studies by Altintas [8,9] on the motion of single dislocations through square arrays of fixed obstacles gives the result

$$\tau_c = \tau_0 + \left[\frac{K_\beta G b}{\sqrt{l_s}}\right]\left[\frac{1}{\sqrt{d}}\right] \tag{12}$$

where T is the line tension of the dislocation, l_s is the mean spacing between obstacles and K_β is a constant that depends on the obstacle strength.

It should be noted that the models described here are not mutually exclusive. They may apply simultaneously, and contribute jointly to the Hall-Petch constant. Furthermore, all models predict a Hall-Petch slope that is proportional to the elastic modulus. Takaki, et al. [10] have summarized the data supporting this result for several metals.

3. Application of the Hall-Petch relation for the strength

The issues that need to be addressed in the application of the classic Hall-Petch relation include the measure of grain size, the use of the mean grain size, and the possible breakdown of the equation when grain size is refined to the nanoscale.

3.1 Selecting the grain size

While the various models described above all lead to a Hall- Petch relation of the classic form, they differ in the appropriate measure of grain size, particularly when the grains are irregular in shape (as in martensitic steels). The dislocation pile-up and the slip models are based on the length of the active slip plane, and would hence use the measure of grain size that determines the mean free path for slip. In the case of irregular grain shapes this should be the mean length of the dominant slip plane ({110} in Fe) in the long dimension of the grain. The dislocation density model relates strength to the grain boundary area per unit volume, and would appear to use a less crystallographic measure of the effective grain diameter. The statistical model would measure the grain size as the width perpendicular to the direction of glide.

In the case of martensitic steel the accepted measure of grain size is the size of the crystallographically coherent block, which may be the packet size or block size, if the packet is subdivided. This grain size is difficult to measure with optical techniques, so the prior austenite grain size is often used instead [11], on the usually reliable assumption that the crystallographic coherence length is determined by the prior austenite grain size, if it is not equal to it. Rapid reversion [12] and rapid solidification [13] treatments have been used to break up the alignment of laths in packets and create ultrafine-grained martensites. In our experience, rapid reversion does not have a dramatic effect on strength, possibly because the laths tend to retain shapes with {110} planes aligned along the long axis. However, rapid solidification was reported to lead to exceptional hardness [13], with a Hall-Petch grain size roughly corresponding to the lath width. It is not clear why this should be the case.

There are also ambiguities in the grain size of steels that have more equiaxed structures. If adjacent grains share (or nearly share) glide planes, they act as a larger unit, despite their apparent independence in optical microscopy. While the application of new technniques in orientation imaging microscopy is making it possible to study orientation distributions and obtain a better picture of the true grain size, very limited data are currently available for steel.

3.2 The mean grain size

Since a material inevitably yields at its weakest element, it may seem inappropriate to use the mean grain size as the constitutive variable in the Hall-Petch relation. Some measure of the maximum grain size, or the least favorable cluster of grains would, arguably, be more appropriate. If, however, the set of samples that are used to test the Hall-Petch relation have microstructures that are geometrically similar, in the sense that they have similar normalized distributions of grain sizes and shapes, then the mean grain size is an appropriate measure. Whatever grain size or grain distribution actually controls the yield mechanism, for geometrically similar microstructures it scales with the mean grain size.

If the mean grain size is a scaling parameter rather than a direct, mechanistic variable then the precise value of the Hall-Petch slope will change with the morphological characteristics of the microstructure. Materials with very different grain size distributions will obey slightly forms of the Hall-Petch relation. This issue has been investigated by Kurzydlowski and Bucki [14], who used powder metallurgy to make aluminum samples with various grain size distributions, and by Weertman and coworkers [15], who studied the behavior of ultrafine grained samples of Cu and Pd as a function of processing. Both sets of investigators found that samples with geometrically different microstructures have significantly different responses to changes in the mean grain size. As expected [14], For given values of the mean grain size, samples with relatively broad size distributions, and, hence, relatively large populations of larger-than-average grains, were softer than those with narrow grain size distributions at the same grain size.

We know of no specific research on the sensitivity of the Hall-Petch slope to the geometric details of the microstructure of steel. However, the similarity in the Hall-Petch slopes obtained by different investigators [10] suggests a fairly weak dependence for the microstructures that are ordinarily encountered.

3.3 Deviations from Hall-Petch at fine grain size

Whatever the mechanism that drives the Hall- Petch relation there should be a grain size so small that it ceases to apply. Research on a variety of materials [16], including steels [17], suggests that this limit falls at about 20 nm mean grain size. Refining the grain size beyond this point does not ordinarily produce higher strength, and may even lead to an "inverse Hall-Petch" behavior in which hardness decreases with finer grain size.

At least three plausible hypotheses have been advanced to explain the small-grain limit of the Hall-Petch relation. First, it is commonly accepted that when the grain size becomes sufficiently small the dominant deformation mechanism will change from transgranular slip to grain boundary sliding, so the Hall-Petch relation no longer applies. While we know of no direct experimental evidence, this hypothesis is supported by recent computer simulations of deformation in ultrafine-grained material [16,18]. However, it is not clear from these simulations that grain boundary sliding could become dominant at grain sizes as large as 20 nm; a recent simulation for Cu suggests a transition at 6-7 nm [18].

Second, very small grains cannot support distributions of dislocations, so the pile-up and dislocation density mechanisms for Hall-Petch behavior cease to apply. Pertinent experimental work has recently been published by Misra, et al. [19], who studied the hardness of thin, laminated films, using transmission electron microscopy to monitor dislocation distributions. They found Hall-Petch behavior in several systems at grain sizes larger than about 20 nm, but significant deviations at smaller sizes. The change in behavior came at approximately the grain size at which the maximum dislocation content in the grains dropped to 2 or less. It should be noted, however, that the geometry of their samples was not conducive to grain boundary sliding.

Third, it is not easy to manufacture ultrafine-grained specimens, and there is a particular risk that the samples that are compared to test the Hall-Petch relation may differ qualitatively in important geometrical features of their microstructures. If the Hall-Petch slope is microstructure-sensitive, as it appears to be in at least some systems, then microstructural differences may produce deviations from the Hall-Petch law. Weertman and coworkers [15] present evidence that this is a source of deviation from the Hall-Petch relation in ultrafine-grained Cu and Pd.

4. The Influence of Grain Size on Fracture

There are at least three distinct fracture modes that are important to the mechanical behavior of steel: ductile fracture by the nucleation and growth of voids, and brittle fracture via transgranular cleavage or intergranular separation. It is not clear that there is a direct grain size effect on ductile fracture, though there is certainly an indirect effect through the effect of grain size on strength. Grain refinement does improve fracture toughness in the brittle intergranular mode. However, I know of no data establishing a constitutive equation of the Hall-Petch type. On the other hand, a Hall-Petch relation of the form of eq. (2) definitely does apply to the fracture via transgranular cleavage.

4.1 Transgranular cleavage and the ductile-brittle transition

In the usual case, transgranular cleavage fracture begins with the initiation of cleavage in a single grain [20]. There is some plastic deformation prior to failure. Initiation often occurs some distance ahead of the crack tip, where the elastic-plastic stress field produces a maximal tensile stress, and at a site of intense local stress concentration. In ferritic or martensitic steel cleavage cracks propagate on {100} planes, which terminate at the grain, packet or block boundaries. If the stress at this point is sufficient to drive the crack through adjacent grains, along {100} cleavage planes within them, then the material fractures in a brittle, cleavage mode. If the stress is insufficient the crack will be stropped at the boundary, and will either be retained as an internal crack or blunted into a ductile rupture void. Since the peak stress at the crack tip increases with the reciprocal root of its size, and the maximum size scales with d, the effective grain size, both the Griffith (energy) and Orowon (stress) criteria suggest that the cleavage fracture stress increases with grain refinement according to the Hall-Petch eq. (2).

In practice, it is difficult to measure the cleavage fracture stress unambiguously. The brittle fracture parameter that is ordinarily measured is the ductile-brittle transition temperature, T_B, and a Hall-Petch relation of the form of eq. (3) has been used to describe its variation with grain size. It is important to recognize, however, that the relation between grain size and T_B is indirect. The mechanism is illustrated in the Yoffee diagram in Fig. 1, where we associate the ductile-brittle transition with the temperature at which the effective yield strength at the crack tip exceeds the brittle fracture stress.

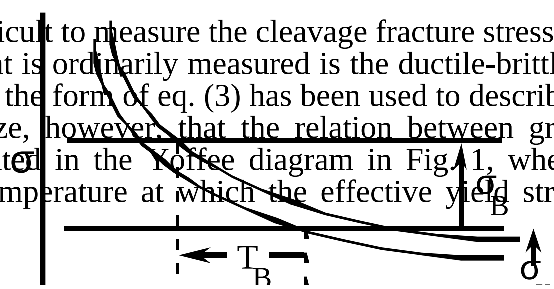

Fig. 1: The Yoffee diagram, illustrating the source of the ductile-brittle transition. σ_B is the critical stress for brittle fracture.

Assuming that the brittle fracture mode below T_B is transgranular cleavage, refining the grain size raises the cleavage fracture stress. This allows the material to have a higher yield strength, since the peak tensile stress in the elastic-plastic field of a crack scales with the yield strength (finite-element calculations suggest that it is 4-5 times s_y for a high strength steel). Some of the increment in cleavage stress must be used to accommodate the higher strength of the fine-grained material. However, for most steels of interest processing to fine grain size causes an increase in cleavage stress that is much greater than the increase in strength, and T_B decreases significantly [21]. If we assume that the increase in the effective yield strength on decreasing temperature is approximately linear, with slope ($d\sigma/dT$), then equations (1) and (2) can be combined to give eq. (3), with the Hall-Petch coefficient,

$$K_B = -\left[\frac{d\sigma}{dT}\right]^{-1}(K_f - K_y) \tag{13}$$

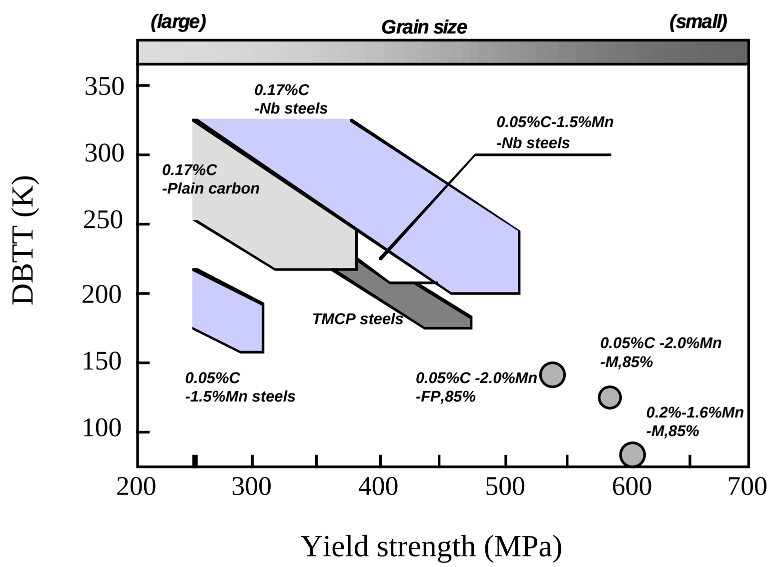

Fig. 2: Increase in the ductile-brittle transition temperature with yield strength on grain refining a variety of common steels [22].

Interestingly, given eq. (1), the Hall-Petch relation for T_B can be re-written

$$T_B = T_0 - \left[\frac{K_B}{K_y}\right](\sigma_y - \sigma_0) \tag{14}$$

which predicts that T_B decreases linearly with the yield strength when grain refinement is the strengthening mechanism. Fig. 2, taken from a recent compilation of data at the Japanese National Institute for Materials Science, shows how well this relation is obeyed for a variety of steels [22].

Prior work in this laboratory [12,20] has shown how advanced heat treatments can be applied to high strength lath martensitic steels to produce ultrafine grain size, and achieve exceptional strength/toughness combinations at cryogenic temperatures. To accomplish this one must recognize that the effective grain size for cleavage fracture in lath martensitic steel is the coherence length along the {100} cleavage plane, which ordinarily traverses martensites packets (or blocks, if the packets are subdivided). Grain refinement is accomplished by controlling the martensitic transformation to break up the crystallographic alignment between adjacent martensite laths, interrupting the cleavage fracture path. In this case, grain refinement does not ordinarily cause a substantial increase in strength, probably because {110} planes lie along the long axis of the laths, which are not significantly refined.

4.2 Hydrogen embrittlement

A somewhat different mechanism applies when lath martensitic steel is embrittled by hydrogen, as in hydrogen charging and in a common mechanism of stress corrosion cracking. If the steel is clean in its grain boundaries, the failure is transgranular, and high-resolution TEM studies have shown that, at least in some typical lath martensitic steels, the mechanism is interlath separation along the {110} lath boundaries [23]. Since the fracture plane is different from that in the cleavage case, the meaning of grain size and the appropriate methods of grain refinement are different as well.

As a specific example, a common heat treatment to lower the ductile-brittle transition temperature of lath martensitic steels is an intercritical temper that introduces thermally stable, precipitated austenite phase along the lath boundaries. The transformation of this austenite during fracture breaks up alignment in the packet and lowers T_B. However, this same treatment *increases* susceptibility to hydrogen embrittlement [24]. The transformation of interlath austenite imposes a wedging load on the lath boundary that helps to split it, promoting early failure.

5. Conclusions

Grain refinement is an effective means for improving the strength and lowering the ductile-brittle transition of structural alloys. The improvement can often be expressed in an equation of the Hall-Petch form. However, the appropriate use of grain refinement requires an understanding of the effective grain size that actually governs the mechanism of yielding or failure.

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