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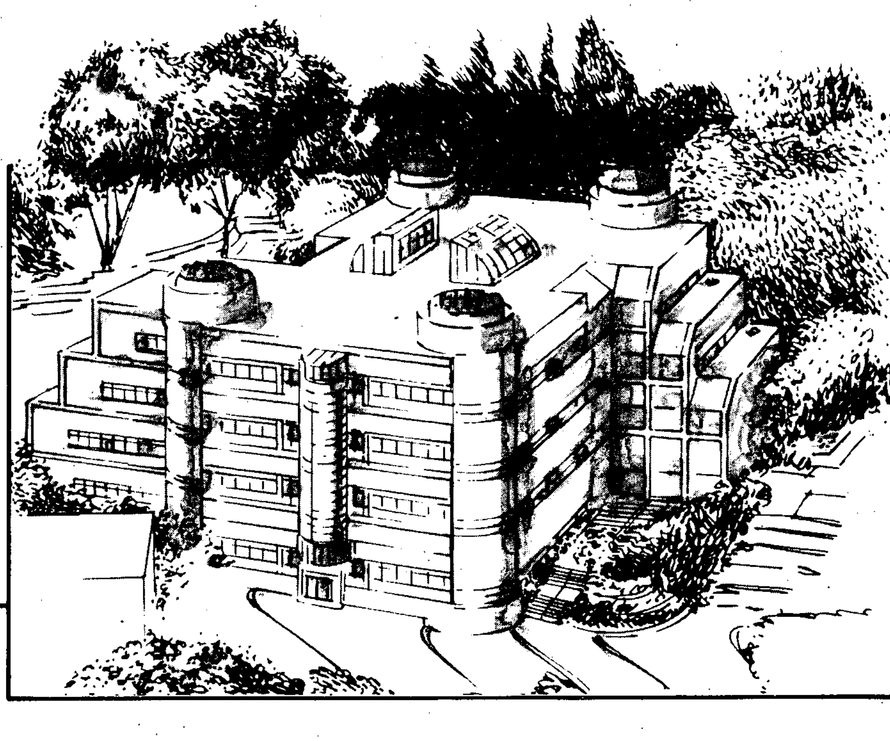

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The Strength and Toughness of Stainless Steels at Cryogenic Temperature

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The mechanical criterion that most often governs the use of stainless steels at cryogenic temperature is the combination of yield strength and fracture toughness. The strength-toughness combination depends on the fracture mode, which should be ductile. The toughness in the ductile mode is affected by the strength, the tensile properties, and the inclusion density. To maximize toughness, the alloy should have a high work hardening rate and a low inclusion count. The toughness of metastable austenitic steels is governed by a balance between stress relaxation by the strain-induced martensitic transformation and the brittleness of the fresh martensite. This balance also governs the response of metastable steels to a high magnetic field; the toughness of relatively stable steels increases when a field is imposed, while that of relatively unstable steels decreases.

I. Introduction

Much of the impetus for the recent development of structural stainless steels for use at cryogenic temperatures comes from the need for strong, tough structural materials for the cases of large, high field superconducting magnets that operate at 4.2K. The wall stresses in such magnets result from the Lorentz force on the conductor inside, and increase geometrically with the size of the magnet and the magnitude of the peak magnetic field. Future engineering devices for applications such as magnetic fusion energy will employ magnets that are several meters in diameter and operate at magnetic fields of 12T or more. These require structural steels with exceptionally high strength at cryogenic temperatures. Other devices, such as high-energy particle accelerators, use finely tuned dipole and quadrupole magnets that require non-magnetic structural steels that combine high strength and toughness with exceptionally low magnetic permeability at 4K.

Most of the new alloy development that has responded to the need for high strength, high toughness cryogenic steels has been done here in Japan [1], and a substantial fraction of the associated metallurgical research has been done here as well. That work is more properly summarized by the active Japanese scientists. I will confine my remarks to a discussion of some of the fundamental issues that concern the mechanical behavior of stainless steels at cryogenic temperature.

The mechanical consideration that most often governs the initial selection of a structural alloy for service at cryogenic temperature is the combination of yield strength, σ_y, and plane strain fracture toughness, K_{Ic}. Both strength and toughness are critical properties since failure may occur through either ductile rupture or fracture. The combination is important since strength and toughness have an inverse relation to one another; an increase in strength at given temperature almost invariably leads to a decrease in fracture toughness. In the design or selection of materials for cryogenic service it is desirable to maximize the strength-toughness combination or, at least, to achieve values that lie within a "design box" in a strength-toughness plot that is bounded by the minimum acceptable strength and toughness values. Since both strength and toughness vary with the temperature the only strictly meaningful design box is one that is defined at the intended service temperature.

There is no reliable quantitative theory of the strength-toughness relation of structural alloys. However, research on the mechanisms of yield and fracture combined with specific studies of the behavior of materials at cryogenic temperatures has produced a qualitative understanding of the low temperature strength-toughness combination that is useful for materials selection, quality control and new alloy design. The following discussion represents our current thinking, and is organized in terms of the mechanisms that may dominate the temperature dependence of the strength-toughness relation: the fracture mode, the tensile properties, and deformation-induced phase transformations.

II. The Fracture Mode

At the micromechanical level the fracture of a material is either ductile, in which case the material is torn apart after considerable local plastic deformation, or brittle, in which case the crack propagates with very little plastic deformation. In most cases there is a first-order correspondence between the level of toughness and the fracture mode: a change from a ductile to a brittle fracture mode causes a substantial drop in the fracture toughness. It follows that the first concern in interpreting the temperature dependence of the strength-toughness characteristic is the possibility of a change in the fracture mode.

The most familiar fracture mode change occurs at the ductile-brittle transition in ferritic steels and other BCC alloys (reviewed in ref. [2]). At high temperature the material fractures in a ductile manner by a microvoid coalescence mechanism and has a relatively high fracture toughness. When the temperature falls below the "ductile-brittle transition temperature", T_B, the mode of crack propagation changes to brittle fracture either by transgranular cleavage of individual grains or intergranular separation along grain boundaries. A ductile-brittle transition is also observed in many FCC alloys, including both austenitic steels [3] and aluminum alloys [4]. In this case the brittle, low-temperature fracture mode is usually intergranular.

The qualitative source of the ductile-brittle transition and its relation to the yield strength can be illustrated by the "Yoffee diagram" shown in Fig. 1, which represents the relative likelihood of plastic deformation and fracture at the tip of a pre-existing crack in a structural material [2]. As the applied stress is increased toward failure the stress at the crack tip reaches one of two levels first: the "yield" stress, σ_Y, at which significant plastic deformation occurs, or the brittle fracture stress, σ_B, at which the crack propagates in a brittle mode by the most favorable mechanism. Extensive plastic deformation at the crack tip limits the local stress and inhibits brittle fracture. Hence the fracture mode is ductile and the toughness high if $\sigma_Y < \sigma_B$. The ductile-brittle transition temperature, T_B, is that at which σ_Y rises above σ_B.

The "yield" stress in the Yoffee diagram is a qualitative concept that is not precisely defined by any available theory. It is most closely related to the yield strength under plane strain conditions, whose value is significantly above the tensile yield strength, σ_y. However, σ_Y varies with σ_y, so the ductile-brittle transition is most pronounced in alloys whose yield strengths increase rapidly at low temperature. The prominent example is carbon steel, in which carbon solutes in the interstices of the BCC structure cause a dramatic rise in strength as temperature is lowered. The ductile-brittle transition is less commonly observed in FCC materials, such as austenitic steels, largely because of the lower increment to the low-temperature yield strength by solute impurities.

As suggested by the Yoffee diagram the fracture mode below T_B is that which provides the smallest fracture stress, σ_B. In BCC material this may be either transgranular cleavage or intergranular separation. In FCC material the brittle mode is ordinarily intergranular. While there are isolated observations of transgranular cleavage in FCC alloys, the cleavage stress is usually high enough that no brittle transition is observed unless an intergranular fracture mode intrudes.

The understanding of the ductile-brittle transition that is gathered in the Yoffee diagram also suggests useful metallurgical mechanisms that can be used to lower or eliminate the ductile-brittle transition. One obvious method is to lower the alloy strength. The low-temperature strength increment can be specifically decreased by removing interstitial solutes or by "gettering" them into relatively innocuous precipitates or second phases. For example, ferritic steels that are intended for cryogenic service are often given intercritical heat treatments that gather carbon into isolated pockets of retained austenite phase or are alloyed with Ti to getter carbon into precipitates [2].

The second obvious method is to raise the brittle fracture stress. The best metallurgical method for doing this depends on the source of the brittle fracture mode. If the fracture is intergranular its source is either a grain boundary contaminant, such as the metalloid impurities S and P in steel, or an inherent weakness of the grain boundary, as is apparently found in Fe-Mn alloys [3] and in many intermetallic compounds [5]. In the case of chemical embrittlement the alloy may be purified of deleterious surfactants, alloyed to getter these into relatively innocuous precipitates, or heat treated to avoid the intermediate temperature regime at which these impurities segregate most strongly to the grain boundaries. When the grain boundaries are inherently weak the metallurgical solution is the addition of beneficial grain boundary surfactants that serve to glue them together. The most prominent of the beneficial surfactants is boron, which is extremely effective in suppressing intergranular fracture in Fe-Mn steels [3] and in Ni_3Al intermetallics [5,6]. Carbon is also an effective surfactant in Fe-Mn steels when it is present in low concentration [3]. When the brittle fracture mode is transgranular, as it is in typical ferritic cryogenic steels, a possible approach is to decrease the effective grain size of the alloy so as to toughen the material by decreasing the mean free path of an element of cleavage fracture. This technique is widely used in the processing and welding of ferritic cryogenic steels [2].

There is a third common method for decreasing the ductile-brittle transition that is less obvious from the Yoffee diagram: processing the material so as to promote delamination perpendicular to the fracture plane that divides the fracture into independent segments that are in nearly plane stress [7]. This technique is ideally equivalent to replacing the plane-strain specimen with a laminate of thin sheets that fracture independently in a nearly plane stress condition. In terms of the Yoffee diagram the effect is to decrease the Yoffee yield strength, σ_Y, at a constant value of the tensile yield strength, σ_y, since the loss of constraint removes the component of stress across the fracture plane that is due to hydrostatic tension. The consequence is that general yielding occurs at the crack tip at a lower value of the total tensile stress across the fracture plane, which is the stress that drives brittle fracture. Processing treatments that achieve delamination have been successfully applied to suppress the ductile-brittle transition in high-strength, low alloy steels, particularly those destined for tankage and pipelines [7]. Delamination may also play an important role in suppressing low-temperature intergranular fracture in some Al-Li alloys [4].

However, it does not follow that delamination treatments necessarily increase the toughness of an alloy in the ductile mode. The metallurgical treatments that induce delamination change the microstructure, weaken it in the short transverse direction, and may liberate a low-energy tearing mode of fracture that is not possible in the monolithic plate. It also does not follow that delamination treatments affect the variation of toughness with temperature in any systematic way. For example, detailed metallographic studies of delamination in the cryogenic fracture of Al-Li alloys have shown that there is no systematic correlation between temperature-induced changes in the level of fracture toughness and changes in the depth or spacing of transverse delaminations [4].

A final comment on the fracture mode concerns metastable austenitic steels, which are FCC alloys that transform to BCC (or BCT) martensite on deformation at low temperature. Many of the most widely used cryogenic structural alloys, such as 304-type stainless steel, are metastable austenites. These materials fracture in a brittle mode evidenced by the predominance of transgranular cleavage on the fracture surface. However, the fracture is preceded by extensive plastic deformation and the toughness is high. The ductility and toughness are a consequence of the phase transformation, whose product is a brittle martensite. The cleavage mode is due to the eventual fracture of the martensite, but the toughness is ordinarily determined by the properties of the strain-induced transformation that precedes fracture. We will discuss these materials in more detail below.

III. Ductile Fracture

The fracture mode that is conducive to a favorable combination of strength and toughness is the ductile mode in which significant plastic deformation precedes fracture. The characteristic variation of the fracture toughness of a ductile material with the yield strength at constant temperature is shown in Fig. 2. Over the intermediate strength range of greatest practical interest the toughness decreases monotonically as the strength is raised.

There are, in fact, several fracture mechanisms that differ in micromechanical detail that are properly called ductile. The mechanism that is most important in stainless steel plate is microvoid coalescence. While there are a number of distinct theories of the microvoid coalescence mechanism of ductile fracture [e.g., refs. 8-10], they have common features and lead to similar qualitative results. The mechanism occurs in two steps. Voids nucleate at inclusions, large precipitates or microstructural flaws, and grow until they join one another. Inclusions, such as oxides and sulfides, are the dominant sources of microvoids in austenitic steels. These create voids through fracture or decohesion from the matrix at relatively low values of the hydrostatic tensile stress that develops in the neck of a tensile specimen and the crack-tip strain field of a specimen that contains a flaw. The juncture of these voids to cause ultimate failure is influenced by work hardening during initial void growth and by fracture or unstable plastic deformation of the matrix material between voids. Most theories assume a regular distribution of voids and predict failure when the stress in the intervening material reaches the critical value for necking or fracture.

For a given inclusion distribution the ductile fracture theories all lead to models of the general form

$$K_{Ic} \propto \varepsilon_f \sqrt{E\sigma_y} \tag{1}$$

where E is Young's modulus, σ_y is the tensile yield strength, and ε_f is the strain to failure, whose precise definition (and power) varies slightly from one model to another. The explicit dependence of the fracture toughness on the yield strength suggests that the two should vary together, in contrast to isothermal toughness data that invariably shows a decrease in toughness as the strength rises (Fig. 2). The resolution of this discrepancy lies in the dependence of the failure strain on the yield strength; ε_f decreases strongly and monotonically with σ_y at constant temperature.

We can gain some insight into the interplay between strength, elongation and work hardening in determining the fracture toughness in the ductile mode by adopting a simple model in which ε_f is proportional to the uniform elongation, or necking strain, ε_c. The strain at which a specimen becomes unstable with respect to necking is determined by the *Considere criterion:*

$$\frac{d\sigma}{d\varepsilon} = \sigma \tag{2}$$

where σ is the true stress and $d\sigma/d\varepsilon$ is the true rate of work hardening. The flow stress, σ, ordinarily increases with the strain while the work hardening rate, $d\sigma/d\varepsilon$, decreases. The necking strain, ε_c, is determined by the point at which the two cross, as illustrated in Fig. 3. Fig. 3(a) illustrates the effect of an increase in yield strength in a material that has a fixed strain hardening behavior. As σ_y increases, ε_c decreases substantially. Given this behavior, equation (1) suggests why the plane strain fracture toughness decreases as the yield strength is raised. Fig. 3(b) illustrates the effect of increasing the strain hardening rate at a given value of the yield strength; ε_c increases as the strain hardening curve is displaced upward (for simplicity, the figure ignores the change in the stress-strain curve due to the increased work hardening). These considerations suggest that ε_f, and, hence, the plane strain fracture toughness, K_{Ic}, in the ductile mode, increase as the yield strength decreases or the work hardening rate increases.

In a typical FCC metal both the strength and the work hardening rate increase as the temperature decreases. The strength rises largely because of the increased effect of solution hardening species; the work hardening rate increases largely because of the difficulty of thermally activated cross-slip at low temperature. The net effect on the fracture toughness depends on the balance between the two; the toughness may rise or fall.

While there is, unfortunately, very little experimental data available to document these trends, two studies seem consistent with them. First, research by Sakamoto, et al. [11] on stable austenitic steels that fractured by microvoid coalescence at cryogenic temperatures showed an improvement in the characteristic variation of impact toughness with strength as the test temperature was decreased to 4 K. Second, systematic measurements of the fracture toughness of Al-Li alloys at cryogenic temperatures have demonstrated an increase in the toughness with increasing tensile elongation with relatively small changes in yield [4].

The second parameter that may significantly influence the toughness of a ductile material is the inclusion density, which determines the density of nucleated microvoids that lead to failure. The ductile fracture theories suggest that a change in the inclusion count at constant values of the tensile properties causes the plane strain fracture toughness to rise according to the relation

$$K_{Ic} \propto \frac{\sigma_y^p}{\sqrt{N_v}} \tag{3}$$

where N_V is the volume density of active inclusions and the exponent (p) is 1/2 or 1, depending on the model. Interestingly, the models predict that the inclusion count has a much stronger influence on the fracture toughness as the yield stress rises, which suggests that the effect should be most apparent at the lowest temperatures and in the highest-strength ductile steels. This prediction is in qualitative agreement with a number of recent observations on the behavior of ductile cryogenic steels, including the exceptional values of fracture toughness that have been obtained in ultraclean, high strength austenitic steels in recent work in Japan [1], and a recent observation of dramatic improvement in the toughness of electron-beam welded austenitic steel at 4K, which is attributed to the reduction in oxygen content during electron-beam welding [12].

IV. Metastable Austenitic Steels

The metastable steels that undergo martensitic transformation at low temperature are exceptional in that they may deform extensively because of the contribution of the martensitic transformation, but eventually fail in a brittle mode through cleavage of the fresh martensite. The best available theories of the "transformation toughening" effect suggest that it is primarily due to the relaxation of the stress at the crack tip by the strain associated with the martensite transformation [13-15]. However, the transformation product is a brittle martensite, and the contribution to the toughness is a balance between the relaxation of the stress at the crack tip and the lower stress intensity required for fracture of the fresh martensite phase. Hence the net effect of transformation on the fracture toughness is a balance of two effects; a moderate degree of transformation increases the toughness while a transformation that is too extensive and too early in the fracture process lowers it.

The extent of the deformation-induced martensite transformation increases as temperature decreases below the critical temperature, M_d, which leads to an increase in the fracture toughness as the temperature drops in most metastable austenitic steels. However, either of two effects can cause a maximum in the toughness of a metastable austenitic steel at some intermediate value of the temperature. First, if the degree of transformation becomes too great then a wide field of brittle martensite forms well ahead of the crack tip. The lower toughness of this martensite product causes a decrease in toughness when the extent of transformation exceeds a critical value. Second, the strain-induced martensitic transformation is often assisted by thermal activation, with the consequence that the extent of transformation at given strain decreases when the temperature is lowered to 4K.

The competition between the beneficial effect of the transformation strain and the deleterious effect of the brittle martensitic product largely governs the influence of a high magnetic field on the cryogenic fracture toughness of stainless steel. When a relatively stable version of 304 stainless is tested at 4K in an 8T magnetic field, the fracture toughness rises as shown in Fig. 4(a) [14]. The increased toughness is associated with an increase in the extent of martensitic transformation at the crack tip. However, when a relatively unstable version of 304 (304L) is tested at 4K in an 8T field, the toughness decreases as shown in Fig. 4(b) [16,17]. In this case the crack tip transformation is so extensive that the brittleness of the fresh martensite dominates the fracture toughness. Stable austenitic steels, such as 310, are relatively unaffected by a high magnetic field.

Finally, a martensitic transformation ordinarily has a beneficial effect on the rate of fatigue crack growth [15]. Crack growth is slowed by the relaxation of the stress field at the crack tip. Hence metastable austenitic stainless steels have relatively low fatigue crack growth rates at cryogenic temperature.

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Figure Captions

1. The Yoffee diagram: the ductile-brittle transition is associated with the rise in the effective yield strength, σ_Y, above the brittle fracture stress, σ_B.

2. The decrease in K_{Ic} with increasing yield strength in a ductile material.

3. The influence of yield strength and work hardening on the necking strain. (a) As σ_y increases, ε_c decreases. (b) As work hardening increases, ε_c increases.

4. The influence of an 8T magnetic field on the 4K fracture toughness of (a) 304 stainless, (b) 304L stainless steel.

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