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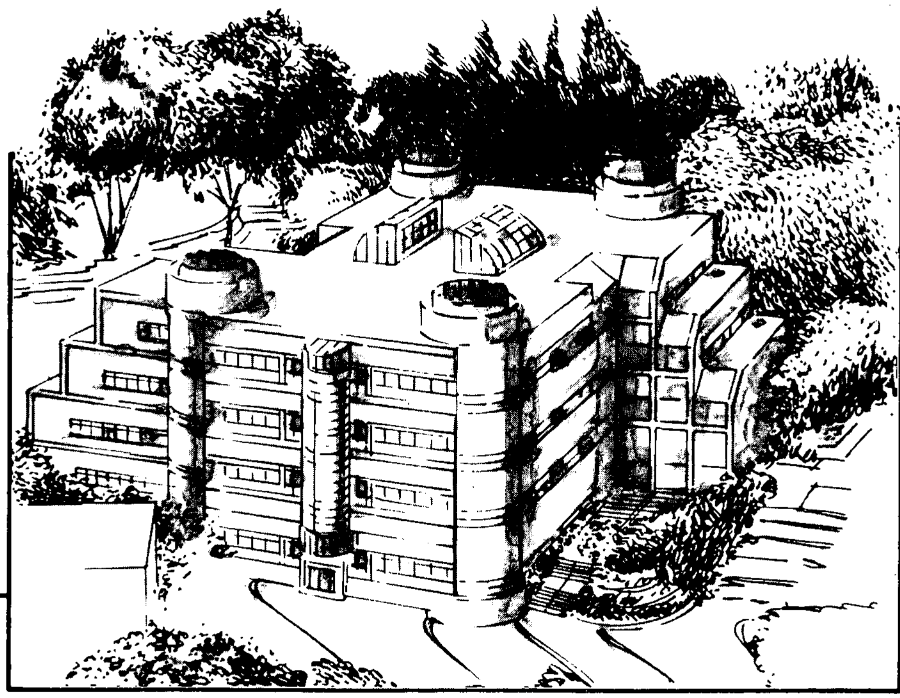

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Cryogenic structural materials can be roughly divided into three classes on the basis of the behavior that determines the strength-toughness characteristic at low temperature. The first class includes materials that undergo a ductile-brittle transition that dominates low-temperature behavior. The second includes materials that remain ductile at all temperatures, whose low-temperature toughness is governed by the interplay of strength and ductility. The third class includes metastable alloys whose ductility and toughness are largely determined by a low-temperature phase transformation. Current models of fracture give some mechanistic insight into the behavior of each class.

I. Introduction

The mechanical consideration that most often governs the initial selection of a structural alloy for service at cryogenic temperature is its strength-toughness combination, its yield strength, σ_y, and plane strain fracture toughness, K_{Ic}. Both strength and toughness are critical properties since failure may occur either through plastic deformation or fracture. The combination is important since strength and toughness have an inverse relation to one another; because an increase in strength at given temperature almost invariably leads to a decrease in fracture toughness, two candidate materials must be compared in toughness at given strength or in strength at given 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. However, cryogenic mechanical tests are difficult and expensive. The mechanical property data base that is available to guide alloy selection or the choice of base compositions for new alloy development is largely confined to properties at ambient temperature. Moreover, economic considerations dictate that at least the initial acceptance tests that are defined for quality control purposes be done at ambient temperature. It is therefore important that the mechanisms that govern the temperature dependence of the strength-toughness combination be known well enough that low-temperature properties can be at least crudely inferred from the metallurgical characteristics of an alloy and its properties at ambient 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. [1]), which is illustrated schematically in Fig. 1. 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 [2] and aluminum alloys [3]. 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. 2, which represents the relative likelihood of plastic deformation and fracture at the tip of a pre-existing crack in a structural material [1]. 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 certainly lies above the tensile yield strength, σ_y, since it corresponds to the plastic flow stress in the presence of the hydrostatic tension at the crack tip. However, σ_Y should change roughly in parallel with σ_y as the temperature is varied. It follows that the ductile-brittle transition should be 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; even interstitial impurities in FCC metals have relatively short-range strain fields because of the size and symmetry of the FCC interstitial sites.

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 [1].

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 and the alkali metals Na and K in Al, or an inherent weakness of the grain boundary, as is apparently found in Fe-Mn alloys [2] and in many intermetallic compounds [4]. 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 [2] and in Ni_3Al intermetallics [4,5]. Carbon is also an effective surfactant in Fe-Mn steels when it is present in low concentration [2]. 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 [1].

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 [6]. 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 [6]. An example is illustrated in Fig. 3 [7]. Delamination may also play an important role in suppressing low-temperature intergranular fracture in some Al-Li alloys [3].

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. For example, the data shown in Fig. 3 are for a steel whose ductile (upper shelf) toughness decreases when the alloy is treated so that it delaminates [7]. 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 [3].

While the description of the ductile-brittle transition given above is applicable to most structural materials, there are situations in which the simple picture of a loss of toughness due to intrusion of a brittle fracture mode at low temperature is clouded or even reversed. For example, impurities such as Na and K in Al alloys and Al, Si and Mg in Be alloys can accumulate in the grain boundaries and form low-melting intermetallics which lead to premature failure in an intergranular mode at temperatures above their melting points [8]. A similar phenomenon is used to make free-machining Al alloys through the addition of Pb and Bi. Decreasing the test temperature suppresses this behavior, and can hence lead to an inverse ductile-brittle transition in which the low-temperature mode is ductile. A more subtle inverse transition has been observed in the Al-Li alloys 2090 [9] and 2091-T6 [3]. In this case the high-temperature mode includes a significant admixture of a fracture that appears to involve transgranular cleavage. At lower temperature the cleavage features disappear; the fracture mode is more ductile and the toughness is much greater. While the source of the cleavage-like fracture is not certain, the available evidence suggests that it is an impurity effect that is associated with rapid diffusion of alkali impurities [9]. At low temperature impurity diffusion is slow and the cleavage-like fracture disappears.

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. 4. 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 plate material, and which has received the greatest theoretical attention, is microvoid coalescence. We shall discuss this mechanism as a prototype for ductile fracture behavior. While there are a number of distinct theories of the microvoid coalescence mechanism of ductile fracture [e.g., refs. 10-12], 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 in steel, are the dominant sources of microvoids in most cases. 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. It is hence often possible to assume the presence of voids. While there are many uncertainties regarding the participation of voids from secondary sites that form later in the fracture process, a simple model is derived by assuming a distribution of voids and assigning a failure criterion that governs their juncture with one another. The failure criterion must somehow account for work hardening during initial void growth and unstable void growth due to fracture or unstable plastic deformation of the matrix material between them. The usual approach is to 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. 4). The resolution of this discrepancy lies in the dependence of the failure strain on the yield strength; ε_f generally decreases strongly and monotonically with σ_y at constant temperature.

When the temperature is lowered, however, equation (1) permits either an increase or a decrease in the fracture toughness, depending on the relative variation of ε_f and σ_y with T. There are countervailing tendencies in the dependence of the failure strain on temperature. These can be generally understood by assuming that the failure strain in eq. (1) scales roughly with the uniform elongation, which is given by the necking criterion,

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

where $d\sigma/d\varepsilon$ is the true work hardening rate and σ is the true stress. The work hardening rate is ordinarily higher at low temperature because thermally activated processes that soften the material are more difficult. This trend increases the failure strain and raises the toughness at given strength. On the other hand, the stress at given strain is higher at low temperature because of the thermal contribution to the yield strength, which tends to decrease ε_f and, hence, the fracture toughness. Balancing these two effects in light of equation (1) suggests a behavior like that shown schematically in Fig. 5. The strength-toughness characteristic curve should be favorably displaced as the temperature decreases since increased work-hardening raises the toughness for given yield. However, the increase in strength as temperature drops causes a decrease in toughness that compensates, and often overwhelms, the increase in toughness due to the better strength-toughness characteristic.

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. [13] 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 [3] (though the fracture mode in this case is not simple microvoid coalescence).

These results suggest that the toughness of a ductile structural alloy will increase as temperature decreases if the work hardening rate is a strong function of temperature while the strength is not. This is the case in some aluminum alloys. The toughness will decrease if the strength is a strong function of temperature, as it is in typical austenitic steels.

The final 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

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

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 [14], 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 [15].

IV. Metastable Austenitic Steels

The metastable steels that undergo martensitic transformation at low temperature are exceptional in that they may undergo extensive elongation 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 [16,17]. 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 a moderate degree of transformation increases the toughness while a transformation that is too extensive and too early in the fracture process decreases 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, the toughness often reaches a maximum at temperature near 77 K, and decreases again if the temperature is dropped further to 4K. There are two possible causes for this effect, which are not clearly distinguished in studies of available metastable austenitic steels. First, there is usually at least some thermal activation required for the strain-induced martensite, which has the consequence that the extent of transformation at given strain decreases as the temperature is dropped below 77 K, resulting in a decrease in toughness. Further, if the transformation is too extensive, that is, if the transformation is "stress-induced", 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.

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Figure Captions

1. Schematic drawing showing the ductile-brittle transition.
2. 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.
3. The variation of Charpy impact energy with temperature in Fe-1Mn steel processed to achieve delamination perpendicular to the fracture plane, showing the drop in T_B and decrease in the upper shelf toughness.
4. The decrease in K_{Ic} with increasing yield strength in a ductile material.
5. Expected variation of the ductile strength-toughness characteristic with temperature.

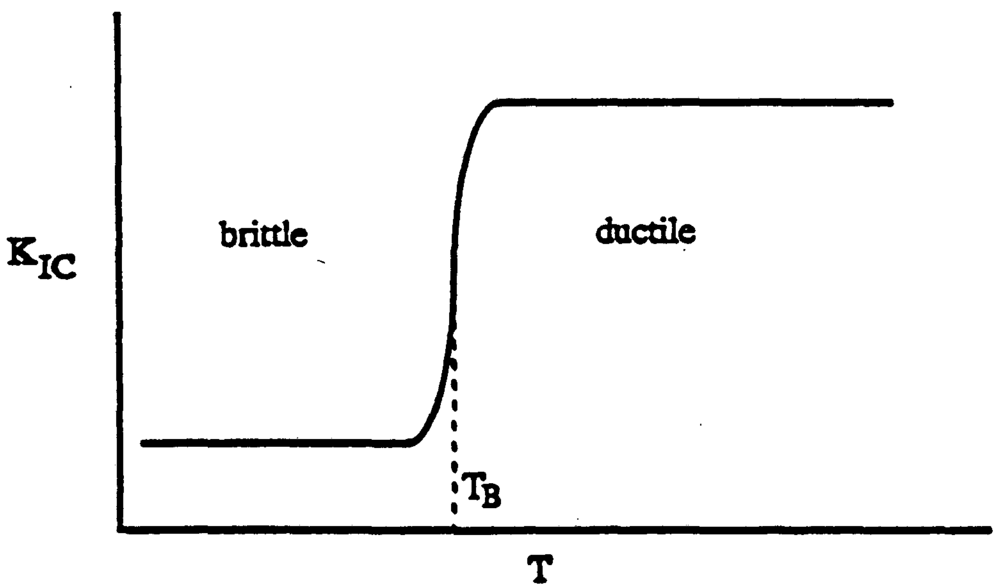


Figure 1

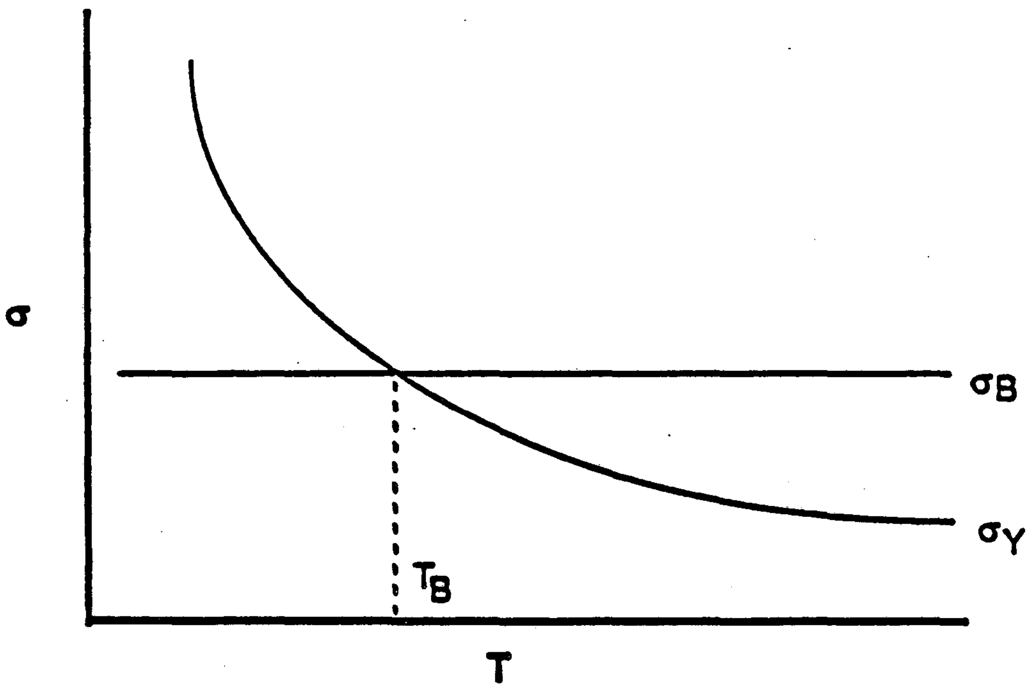
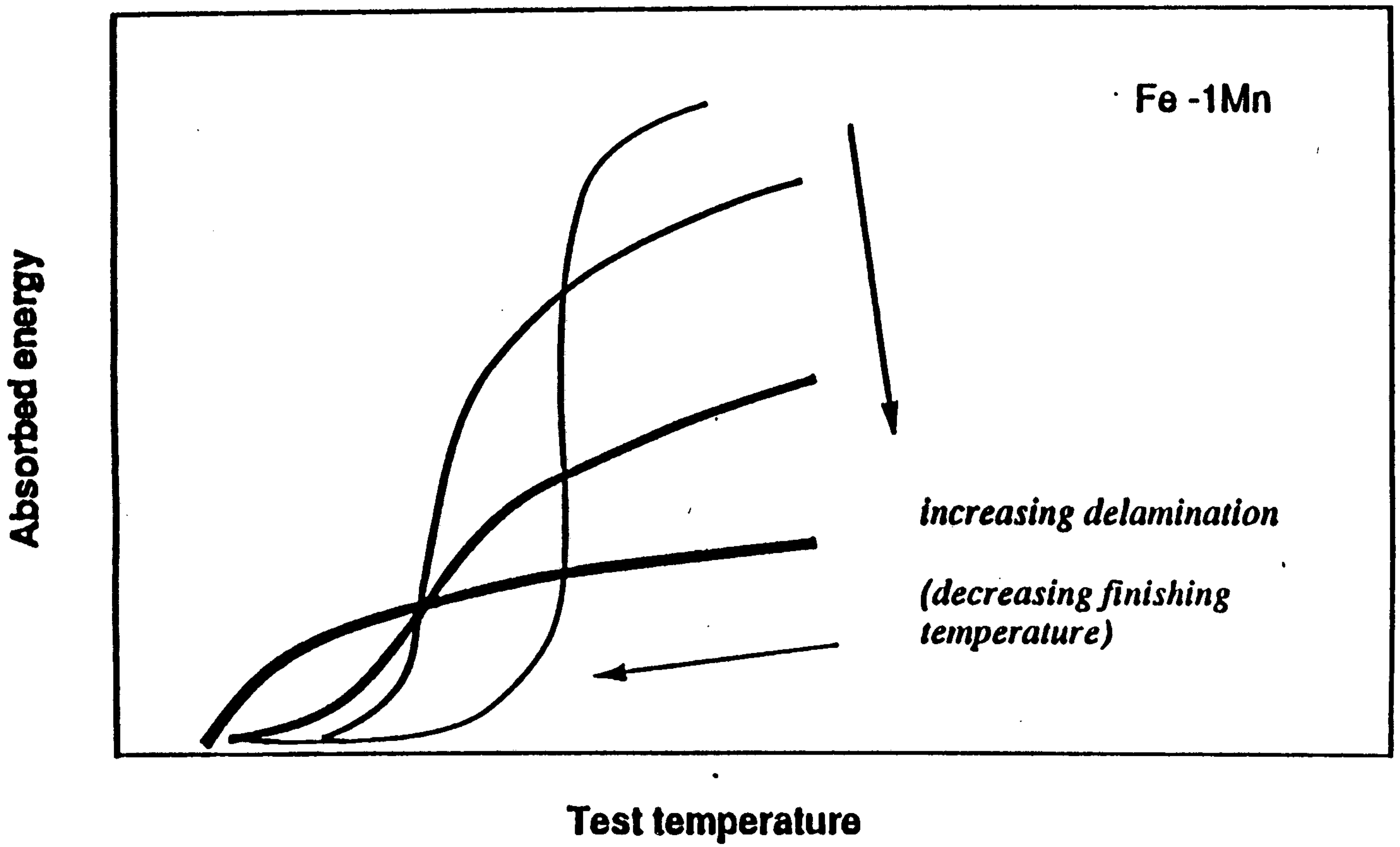


Figure 2

Effect of intergranular delamination on Charpy impact energy

(crack divider orientation)



Bramfitt and Marder, 1977
Met. Trans., 8A:1263

Figure 3

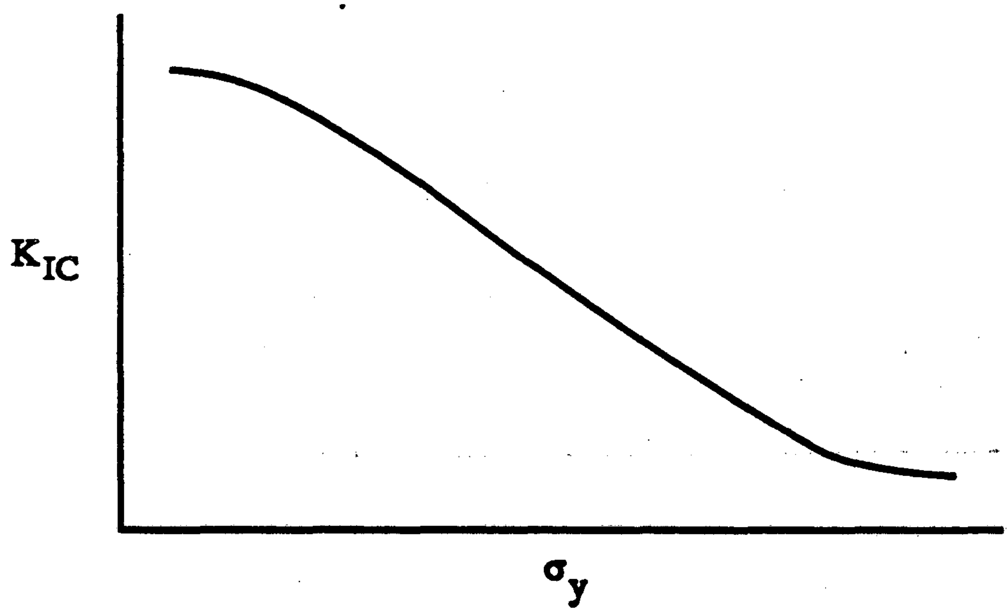


Figure 4

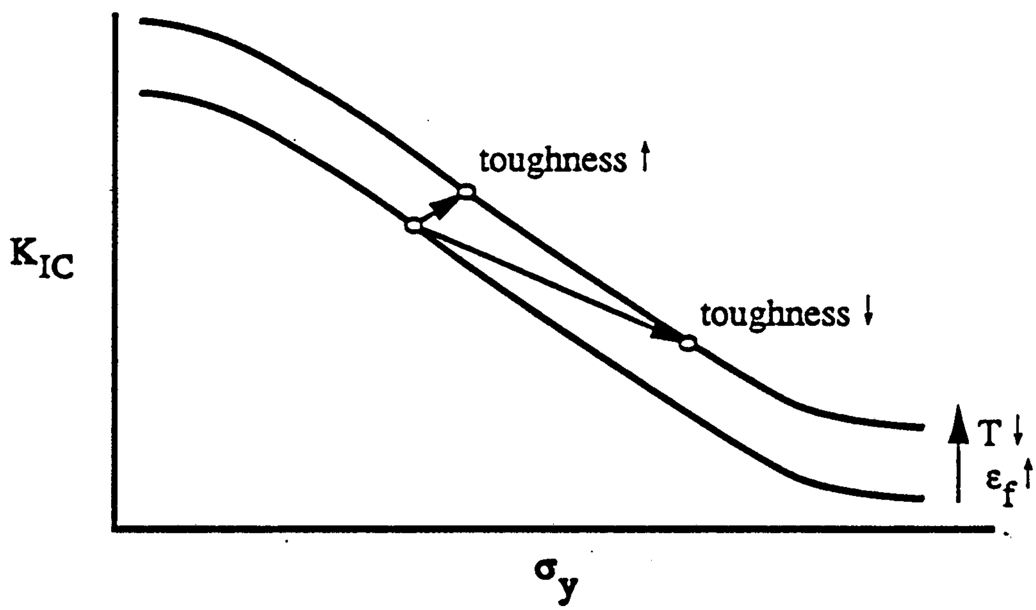


Figure 5

References

1. J. W. Morris, Jr., Adv. Cryogenic Eng., 32, 1 (1985)

2. M.J. Strum, S.K. Hwang and J. W. Morris, Jr., in *Interfacial Structure, Properties and Design*, MRS Symposium Volume 122, 1988, p. 467; M.J. Strum, PhD Thesis, Dept. Materials Science, Univ. California, Berkeley, 1988

3. *Al-Li Alloys*, Williamsburg, Va., 1989; J. Glazer, PhD Thesis, Dept. Materials Science, Univ. of California, Berkeley, 1989

4. C. T. Liu, C. A. White and J. Horton, Acta Met., 33, 213 (1985)

5. K.-M.Chang, S. C. Huang, A. I. Taub, G.-M. Chang and J.W. Morris, Jr., Met Trans, 18A, 1819 (1987)

6. I. Tamura, C. Ouchi, T. Tanaka and H. Sekine, *Thermomechanical Processing of High-Strength, Low-Alloy Steels*, Butterworths, London 1988, pp. 101-116

7. B.L. Bramfitt and A.R. Marder, Met. Trans., 8A, 1263 (1977)

8. D. Webster, Met. Trans., 6A, 803 (1975); 18A, 2181 (1987)

9. D.N. Frager, M.V. Hyatt and H.T. Diep, Scripta Met., 20, 1159 (1986)

10. G.T. Hahn and A.R. Rosenfeld, *ASTM STP 432*, American Society for Testing and Materials, Philadelphia, 1968, p. 5

11. P.F. Thomason, Int. J. Fracture Mech., 7, 409 (1971)

12. G.G. Garrett and J.F. Knott, Met. Trans., 9A, 1187 (1978)

13. T. Sakamoto, Y. Nakagawa, I. Yamauchi, T. Zaizen, H. Nakajima and S. Shimamoto, Adv. Cryo. Eng., 30, 137 (1984)

14. H. Nakajima, et al., Adv. Cryo. Eng., 32, 347 (1986)

15. T. Ogawa, Nippon Steel, Private Communication, 1989

16. A.G. Evans and R.M. Cannon, Acta Met., 34, 761 (1986)

17. J.W. Chan, Z. Mei, J. Glazer, P.A. Kramer and J.W. Morris, Jr., Acta Met., (in press)

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