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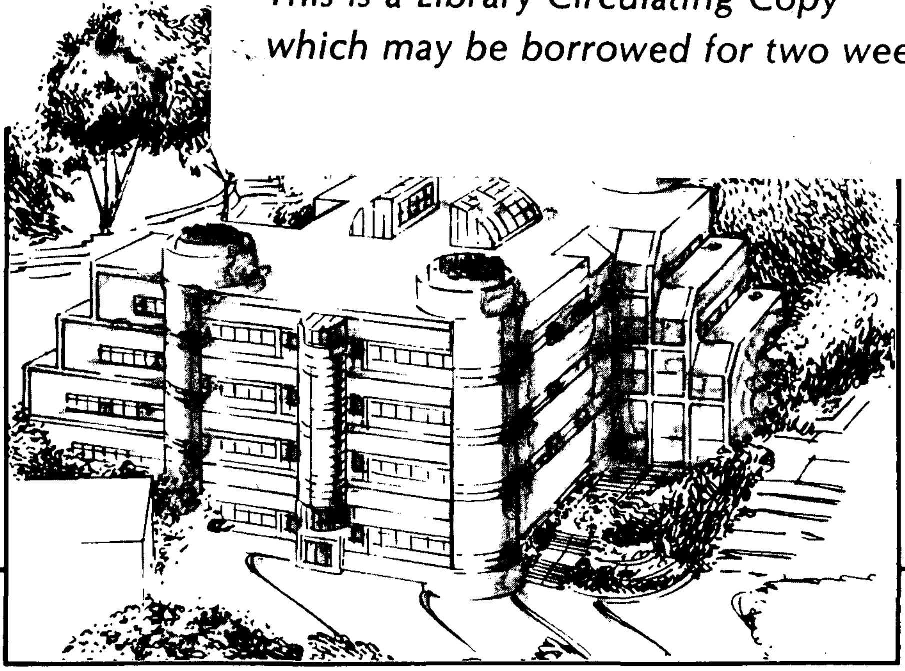

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Mechanical Behavior of High-Strength Aluminum Alloys at Cryogenic Temperatures

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This paper reviews the cryogenic mechanical behavior of high-strength aluminum alloys and describes some of the potential applications of these alloys at cryogenic temperatures. The aluminum alloys are unusual in that their mechanical properties often improve with decreasing temperature. The mechanical property data is summarized and briefly compared to that for other f.c.c. materials, such as austenitic steels and nickel-based alloys, many of which also have better mechanical properties at low temperature. The cryogenic properties of aluminum alloys are discussed and interpreted in light of the current mechanistic theories of the influence of temperature on mechanical behavior. The discussion emphasizes the behavior of advanced aluminum-lithium alloys, which have exceptional mechanical properties at cryogenic temperature.

Introduction

Applications

Interest in the cryogenic properties of aluminum alloys has, historically, been driven by specific technological needs that have arisen, for example, in the development of transportation systems for liquified natural gas (LNG) and cryogenic tankage for spacecraft. As a consequence, the published information on the cryogenic properties of commercial alloys consists almost exclusively of tabulations of mechanical properties with little or no discussion of microstructure or mechanisms. The work includes a considerable effort in the early 1970's on extremely tough, low-to-moderate strength alloys such as 5083-O (nominal composition, in weight percent, Al-4.4Mg-0.7Mn), since these were of interest for use in LNG (111 K) [1-4]. A second body of work was spurred by the development of the space shuttle, whose external tank is constructed of 2219-T87 (nominally Al-6.4Cu). Both applications involve welded cryogenic tanks, and hence stimulated research on the weldability of aluminum alloys for cryogenic use [5-6]. Much of the available data is summarized in ref. [1], which includes cryogenic mechanical properties for most commercial alloys that have been measured, although 4 K toughness data remains scarce. More fundamental research on aluminum alloys at low temperature has addressed their tensile properties almost exclusively, and has focussed on simple alloys that have very little behavioral similarity to the commercial alloys of technological importance.

The last three years have seen a resurgence of research on the cryogenic properties of high strength aluminum alloys that is driven by the simultaneous appearance of new programs in advanced aerospace systems and new high strength aluminum alloys with particularly promising properties. The most important of the new alloys are the advanced Al-Li alloys that have been recently commercialized in the United States and Europe [7-8], which offer 7-10% lower density and ≈10% higher elastic stiffness than the best available aerospace alloys at comparable levels of strength and toughness. Moreover, early work on the Al-Cu-Li alloy 2090-T81 (Al-2.7Cu-2.2Li-0.1Zr) showed that the fracture toughness of this alloy improves dramatically as temperature decreases [9-10]; the strength- toughness combination at 4-77 K is substantially superior to that of any other commercially available aluminum alloy. The important potential applications include cryogenic tankage for advanced aerospace systems and cryogenic structures for high energy physics devices.

Aerospace cryogenic tankage. The aerospace cryogenic structures of greatest current interest include expendable tanks for space vehicles such as the space shuttle, which are currently made of alloy 2219-T87, and integral tanks for future hypersonic aircraft. The fuels in both cases are liquid hydrogen (20 K) and liquid oxygen (100 K). The alloys for expendable, external tanks require high strength and toughness at 20 K, low density, and high stiffness. The alloys must be weldable, and, because of the need for low overall weight, must be formable or machineable into relatively complex shapes. Detailed property requirements are being developed as part of the NASA Advanced Launch System (ALS) program in the United States. The properties required for integral tankage in hypersonic vehicles depend on whether the tank is parasitic or part of the aircraft structure. Alloy strength is more important in the latter case. Current design activities in hypersonic vehicles include the National Aerospace Plane (NASP) in the United States and HOTOL in Europe [11-12]. Japan is also studying the development of a space plane, the Hope or H2 orbiting plane.

High energy physics devices. Advanced accelerators and magnetic fusion energy devices operate at 4 K so that they can use large superconducting magnets. The magnet case materials must have good mechanical properties at cryogenic temperature. Because of its physical properties (e.g. coefficient of thermal expansion) and light weight aluminum is sometimes selected over much higher strength cryogenic steels. An example is the Nb_3Sn force-cooled conductor magnet built by Westinghouse for the Large Coil program in the US [13], which had an external case made of alloy 2219-T87. Aluminum alloys are now being considered for magnet collars for the Superconducting Supercollider (SSC) in the US. These may be made of 7075-T6 or 2090-T81 (for strength). Potential applications at the European high energy physics facility at CERN include shrink-fitted rings and collars of alloys 2014 and 5083. Cryogenic aluminum alloys may also be used in "radiation-thin" superconducting detector magnets and beam vacuum chambers [14-15]. These designs require high strength, moderate toughness and good fatigue resistance.

Among the other past and potential cryogenic applications for aluminum alloys are LNG tankers, cryogenic wind tunnels and missiles.

Mechanical properties and manufacturability

Relatively little fundamental research has been done on the cryogenic mechanical behavior of high-strength aluminum alloys, and none of the commercial alloys now in use were designed for low temperature performance. The most widely utilized high-strength cryogenic alloy, 2219-T87, was designed for high temperature service. The most promising of the current alloys, the Al-Li alloy 2090, was designed to be a replacement for 7075 in aircraft structures. However, the important new applications described above have stimulated new research on the mechanisms of cryogenic behavior [16-23] and the design of aluminum alloys that are specifically intended for cryogenic service [24].

Mechanical properties. The most striking aspect of the cryogenic mechanical behavior of 2219-T87 and 2090-T81 is the dramatic improvement in the strength-toughness combination with decreasing temperature. This behavior is atypical, even for aluminum alloys. Understanding the cryogenic behavior of high-strength aluminum alloys, and especially aluminum-lithium alloys, is the central problem limiting alloy design for cryogenic service.

Manufacturability. Because of the increasingly high premium placed on lowering launch costs for aerospace vehicles, manufacturability issues have taken on great importance. Most of the work oriented toward cryogenic applications has been focussed in two areas: superplastic forming and welding.

Superplastic forming is an appealing manufacturing technology because it allows production of complex shapes with a minimum expenditure of material, energy and labor. Several aluminum alloys are currently available in superplastic modifications, including 2090 and other aluminum-lithium alloys. The cryogenic properties of superplastic 2090 sheet peak-aged after forming have been examined and are sufficiently promising to encourage further research [25,26]. Like the standard plate material, the tensile properties of the material improve with decreasing temperature; however, the toughness decreases somewhat. Other aluminum-lithium alloys display good superplastic formability [27], but are relatively brittle at low temperature [26].

Good weldability is an essential prerequisite for any alloy employed in cryogenic service. The alloy 2219 has been used extensively for cryogenic applications in large part because of its good weldability [5]. Of the new aluminum-lithium alloys, most attention has been given to alloy 2090 which has also been shown to be weldable if proper pre-cleaning procedures are used and when post-weld aging is feasible. Characterization of the mechanical properties of the welds at low temperatures has also been conducted [28,29].

Mechanical behavior of high-strength aluminum alloys

This portion of the paper is broken up into four sections each of which discusses the variation with temperature of a particular mechanical property: the elastic modulus, yield and ultimate tensile strengths, tensile elongation and fracture toughness. This section will focus primarily on surveying the available data for aluminum alloys, with an emphasis on aluminum-lithium alloys; a more detailed discussion of mechanisms is deferred to the next section.

An examination of the literature suggests the behavior of aluminum alloys is a subset of the more general problem of generating a unified picture of the mechanical behavior at low temperatures of f.c.c. alloys in general. Austenitic steels and nickel-based alloys also show a wide range of low temperature behavior ranging from much improved to much deteriorated from their room temperature properties. It is likely that the mechanisms of improved low temperature behavior are common to all of these alloy systems. For this reason some comparisons are drawn between aluminum and other f.c.c. alloys. Because the f.c.c. alloys have widely disparate strengths, comparisons between them are difficult. Because the shear modulus G is a scaling factor in theoretical quantities related to strength, the data should be scaled by G or the elastic modulus E. Relative to their stiffness, high strength commercial aluminum alloys are as strong as the highest strength steels. An aluminum alloy with a yield strength of 80 ksi (550 MPa) is equivalent to a steel with a strength of 200 ksi (1380 MPa).

It is difficult to find both tensile and toughness in the literature across the temperature range 300 K to 4 K for most alloys, and even more unusual to find accompanying microstructural and fractographic information. It is hoped that this review will stimulate interest in developing this kind of data.

Elastic properties

The variation of elastic properties with temperature is one of the few area of mechanical behavior that has been well characterized and is at least partially understood. Aluminum alloys show an increasing elastic modulus and decreasing Poisson's ratio with decreasing temperature. The change in these properties relative to their values at room temperature appears to be constant for all aluminum alloys even though their room temperature elastic properties are different (e.g. Al-Li alloys are 7-10% stiffer than conventional Al alloys). A compilation of elastic properties of cryogenic structural materials has been published by Ledbetter [30]. With the exception of Invar (36Ni-64Fe), all of these increase in stiffness by a similar percentage as aluminum alloys.

Strength

The variation in yield strength and ultimate tensile strength with temperature has been documented for a number of alloys in the range 300-4 K. The variation in yield strength with temperature for a selection of f.c.c. alloys is illustrated in figure 1 [31]. However, most of the low temperature data is for a few discrete temperatures: at 4 K (LHe), 20 K (LH$_2$), 77

K (LN_2) and 200 K (dry ice in alcohol). A few studies have examined much larger number of temperatures. The yield and ultimate strengths generally increase with decreasing temperature, but the detailed studies have indicated that they are not necessarily smoothly varying (e.g. [22]). In addition L, LT strength anisotropies are not necessarily maintained through this temperature range and may even reverse. This is perhaps particularly surprising with respect to yield strengths. Ultimate tensile strength variations are more easily explained in the sense that work hardening is a complicated phenomenon, influenced by many factors that may have different temperature dependences (e.g. ease of cross-slip, which depends on texture). This effect is quite strong in 2090 and 2091 (Al-2.2Cu-2.0Li-1.5Mg-0.1Zr), both of which have a relatively anisotropic grain structure and are strongly textured.

Fracture toughness

In view of the fact that strength increases with decreasing temperature, it would perhaps be most easily explicable if the toughness decreased with decreasing temperature along the same strength-toughness relation that holds for the material at room temperature. This type of behavior has been observed in Fe-Mn alloys [32]. It might be expected that this would be the case if the fracture mode remained unchanged, but that a dramatic decrease in toughness would be observed if it did change to a lower energy mode (i.e. a ductile-brittle transition such is observed in many b.c.c. steels). However, this is not the case for 2219 and 2090 or for a number of steels and nickel alloys which remain ductile to 4 K. In fact, upper shelf toughness can increase with decreasing temperature and increasing strength in alloys which do show a ductile-brittle transition [33]. The mechanisms behind this behavior in aluminum are currently under study and will be discussed in considerable detail below.

The strength-toughness combination for aluminum alloys 2090 and 2219 is illustrated in Figure 2. The principal source of the improvement is the substantial increase in fracture toughness at low temperature; the yield strength is only slightly affected. As Figure 3 illustrates, this behavior is atypical, even for aluminum alloys. Other alloys show either an improvement in strength at relatively constant toughness or a deterioration in the toughness as the temperature drops. Both 2219 and 2090 also have improved tensile elongation at low temperature. Although 2219 has been used in cryogenic applications for some time little is known about the mechanism behind its improved properties at low temperature. Similarly, although the behavior of 2090 is now under study by several groups, the beneficial mechanisms have not yet been clarified so that they can be phrased in way that is useful for alloy design. The improvement is not simply due to the presence of lithium, since other aluminum-lithium alloys do not show improvements as striking as those in 2090 [26].

Elongation

Although tensile elongation is dependent on stress-state it represents an important point on the forming limit curve. As shown in Figure 4, many alloys (2000 series aluminum alloys, OFHC copper, etc.) show an increase in tensile elongation at low temperature which may be a reflection of the increased strain hardening rate that can be maintained at low temperature. Not all of these alloys show improved toughness, but many of them do. Again it makes sense to distinguish between those alloys that show a change in failure mode and those that do not (e.g. necking or not).

Mechanisms

This section describes the current theories for improved mechanical behavior in aluminum alloys at low temperature. The discussion is heavily weighted toward aluminum-lithium alloys as there have been relatively few studies that combined mechanical testing with microstructural analysis, particularly with respect to fracture toughness. However, these mechanisms should also be judged on their ability to explain the behavior of other aluminum alloys and f.c.c. metal alloys in general. Accordingly, some comments on the behavior of austenitic steels and nickel alloys are provided for comparison

Sources of improved toughness at low temperature

Influence of strength and strain hardening rate on J_{Ic}. The yield strength, ultimate tensile strength and strain hardening rate all increase with decreasing temperature in 2090 and many other aluminum alloys. Analytic theories of elastic-plastic fracture predict that if the fracture mode is unchanged, these increases should result in an increased value of the J_{Ic}. For example, if we assume strain-controlled fracture and that the relevant microstructural parameters are constant,

$$J_{Ic} \propto \left(\varepsilon_f^*\right)^{n+1} \sigma_y b \tag{1}$$

where ε_f^* is the true strain to fracture, n is the strain hardening rate ($\sigma = k\varepsilon^n$), σ_y is the yield strength and b is the Burgers vector (inserted for dimensionality). For aluminum-lithium alloys the few available data points appear to fit this relation reasonably well if the further assumption is made that the fracture strain corresponds to the tensile strain to fracture; however, there is not enough data to warrant a stronger conclusion than that continued research along this path is worthwhile.

Clearly, if the fracture mode changes, this argument is invalid. An example is 2090 thermomechanically-processed to be superplastically formable and peak-aged. The strength and strain-hardening rate of this material increase with decreasing temperature, while the toughness declines. However, the fracture mode, which is ductile shear at room temperature, becomes increasingly intergranular at low temperature [25].

Intergranular delamination. Intergranular splitting (delamination) along LT planes is a relatively common observation in aluminum alloys fractured at low temperature. It has been observed in 7075 [23], 2090 [10,18,26] and various Al-Cu-Li-Mg-Zr alloys including 8090 [18,19,22]. In general, both the number and depth of the cracks increases with decreasing temperature. With the exception of Saji and Verzasconi, none of these investigators performed tests at temperatures below 77 K. A typical fracture surface and matching crack profile are shown in Figure 5.

The influence of this laminar cracking on the fracture toughness remains in question. Dorward first proposed that the improved toughness of 2090-T81 at low temperature could be attributed to increasing intergranular splitting which effectively places the crack in plane stress and thus increases the apparent fracture toughness [10]. This explanation is consistent with the slight decrease in S-L fracture toughness he observed; in this orientation the crack runs along a plane which splits easily. This view is also supported by Rao et al. [18] who have done extensive studies of the fracture behavior of several aluminum-lithium alloys at low temperature. However, other investigators disagree with this viewpoint. Niinomi et al. note that in previous work on 7N01 and 5083, laminar cracking was associated with a decrease in toughness with decreasing temperature [22]. Saji et al. did a detailed study of the tensile behavior of 7075 between 6.5 K and room temperature. They report increasing toughness (as estimated by the work to fracture) and elongation with decreasing temperature until a peak at 30 K and decreasing values from there to 4 K. The peak in toughness and elongation is associated with a maximum amount of laminar splitting and a minimum in the localized (post-uniform) strain. However, previous direct measurements of the toughness have shown that the toughness of 7075 decreases between room temperature and 77 K [23]. Thus, while there is considerable evidence for increased laminar cracking at low temperatures in aluminum alloys, it is not clear that it is presence is responsible for increased low temperature toughness or even that it is always associated with increased toughness at low temperatures.

Of the authors above, only Dew-Hughes and Niinomi provide an explanation for the increase in intergranular splitting at low temperature. Both authors believe that the increase in cracking is due to the increased work hardening ability of the matrix, which increases the strength of the matrix relative to the grain boundaries.

Liquid metal embrittlement. Webster [20] has proposed that low melting point phases lower the toughness of these alloys at room temperature from the values at lower temperature. He cites phase diagrams for Al with Na, K, and rare-earths which contain liquid phases down to about 195 K. Apparently, the presence of the liquid phase at grain boundaries and grain boundary triple points results in liquid metal embrittlement. At lower temperatures these phases freeze out and the toughness increases. Niinomi et al. support this interpretation based on their observation of Na and K segregation on the fracture surface at room temperature and 123 K but not at 77 K. They compare the expected temperature at the crack tip after accounting for adiabatic heating to Webster's phase diagrams and find good agreement between these temperatures. There are several important flaws in this argument. The first is that the temperature effect does not appear to be significantly decreased in alloys prepared from extremely pure starting materials. The second is that the melting points of all of the proposed liquid phases are above 77 K, so this theory cannot be used to explain the continued increase in toughness between 77 and 4 K measured for 2090. Thirdly, it is not clear that the gross fracture mode remains unchanged.

Others. The mechanistic theories described above are not necessarily mutually exclusive or a complete list. Strain hardening and intergranular delamination are probably closely related. Texture and grain size and shape are almost certainly important.

Sources of improved elongation at low temperature

Many aluminum-lithium alloys apparently fracture without necking, yet show increased elongations at low temperature. However, analysis of the tensile true stress- true strain curve can be used to show that failure occurs almost exactly at the instability point at which localized deformation begins. This observation is best illustrated with a Considere plot which compares the true stress to the instantaneous strain hardening rate as a function of true strain. A brittle material fractures without meeting this criterion, whereas a ductile one fractures at or beyond it. Tensile instability (necking) occurs when the criterion

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

where σ is the true stress and ε is the true strain. Figure 6 shows Considere plots for 2090-T81 aluminum-lithium alloy tested at three different temperatures. Only one specimen showed observable necking, but all failed after the Considere criterion was satisfied. It can be seen from the plots that the increased tensile elongation at low temperature is associated with the increased strain hardening rate which postpones the tensile instability point to larger strains.

There has been considerable effort on the source of the increased strain hardening rate at low temperatures in relatively simple (and generally low strength) laboratory alloys. It is clear that cross-slip becomes more difficult so that the dislocations become more densely tangled during the deformation process. However, the magnitude of the effect seems to vary from one alloy to the next and must depend on precipitates, texture and other factors. Analysis of data for 2090-T81 and 2090-T4 using the methods described by Mecking [34] for polycrystalline pure metals indicates that extant theory does not apply simply to more complex alloys that are both solute- and precipitation-hardened. Rather than superimposing for different test temperatures early in the deformation process, they are translated from one another by an amount related closely to the yield stress. The analysis does suggest that Stage II (pure hardening) behavior is almost non-existent even at very low temperatures. The plots appear characteristic of Stage III.

Strengthening mechanisms at low temperatures

Although strengthening theory is much more advanced than the theory of other mechanical properties, some questions still remain. Figure 7 shows the variation of yield strength with temperature of 2091-T8 for L and LT orientations [35]. The strength anisotropy varies significantly with temperature. This variation is not easily explained with current theories. Most theories for strengthening by strong obstacles assume athermal glide, i.e. a temperature of 0 K. On the other hand, the strong temperature dependence of hardening by weak obstacles such as solute atoms is well known. It seems plausible that most of the strength difference between room temperature and 4 K is due to solute hardening, but it is difficult to see why the increase in strength should be orientation-dependent. Obviously further work on the temperature dependence of precipitate strengthening at low homologous temperatures is required.

Summary

Research on the cryogenic mechanical properties of aluminum alloys is being conducted at an accelerating pace. The size and nature of the applications now on the horizon has encouraged mechanistic studies that should eventually provide the information required for the design of improved cryogenic alloys. However, current understanding lies far short of that goal. Further work on the reasons for improved strength, toughness and elongation at low temperatures is required. As much as possible, this work should attempt to place aluminum alloys in the context of other f.c.c. alloys with similar relative strength levels.

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Figure Captions

Fig. 1. Variation of yield strength with temperature for a selection of f.c.c. alloys.

Fig. 2. The strength-toughness-temperature relation of the aluminum alloys 2090-T81 and 2219-T87. The strength-toughness trend line for advanced aerospace aluminum alloys at room temperature is shown for comparison.

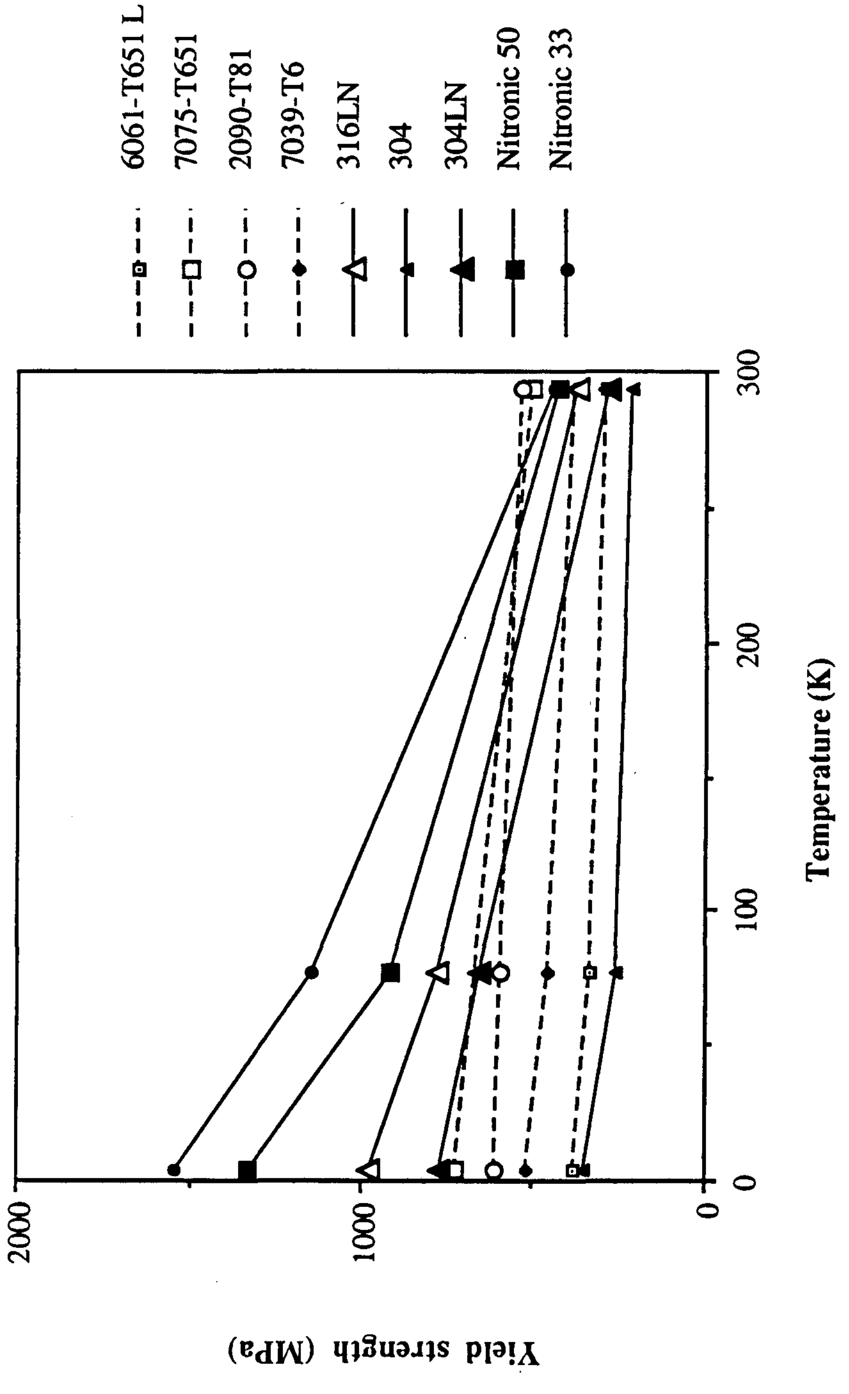
Fig. 3. Fracture toughness relative to toughness at room temperature plotted as a function of temperature for selected f.c.c. alloys. All specimens were in the L-T orientation unless specified otherwise.

Fig. 4. Tensile elongation relative to room temperature tensile elongation as a function of test temperature for some f.c.c. alloys. All specimens were in the L orientation unless otherwise specified.

Fig. 5. (Left) Fracture surface of 2090-T81 J_{Ic} specimen (L-T orientation) broken at 4 K. The fracture appearance is identical to those of specimens tested at room temperature and 77 K. (Right) Profile of the fracture surface shown at left illustrating the intergranular delamination perpendicular to the crack path. The delaminations are deeper and more frequent in samples broken at low temperature.

Fig. 6. Considere plot for 2090-T81 in the L orientation broken at three test temperatures. Specimens were taken from 12.5 cm (0.5 in) plate at quarter-thickness. The strain hardening rate $d\sigma/d\varepsilon$ and the true stress σ are plotted on the same axis because they have the same units.

Fig.7. Influence of temperature on yield strength anisotropy between L and LT orientations with respect to the rolling direction in 3.8 cm (1.5 in) thick 2091 plate. Specimens were taken at quarter-thickness and tested in the stretched and peak-aged T8 condition.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7075-T651
2090-T81
7039-T6
316LN
304
304LN
Nitronic 50
Nitronic 33
Yield strength (MPa)
2000
1000
0
Temperature (K)
0
100
200
300

Figure 1

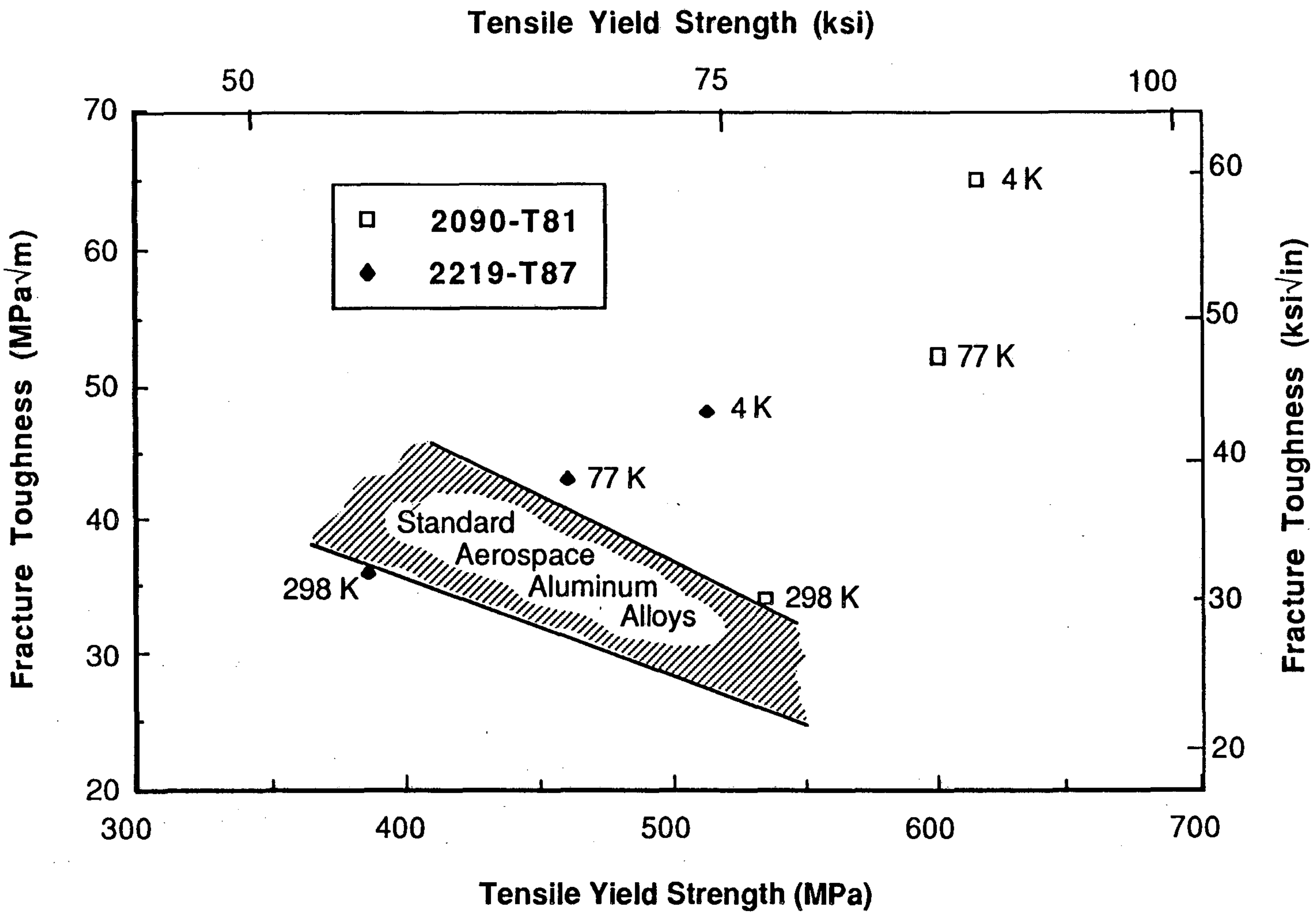

Tensile Yield Strength (ksi)
50
75
100
70
60
50
40
30
20
60
50
40
30
20
Fracture Toughness (MPa√m)
Fracture Toughness (ksi√in)
□ 2090-T81
◆ 2219-T87
4 K
77 K
4 K
77 K
298 K
298 K
Standard Aerospace Aluminum Alloys
300
400
500
600
700
Tensile Yield Strength (MPa)

Figure 2

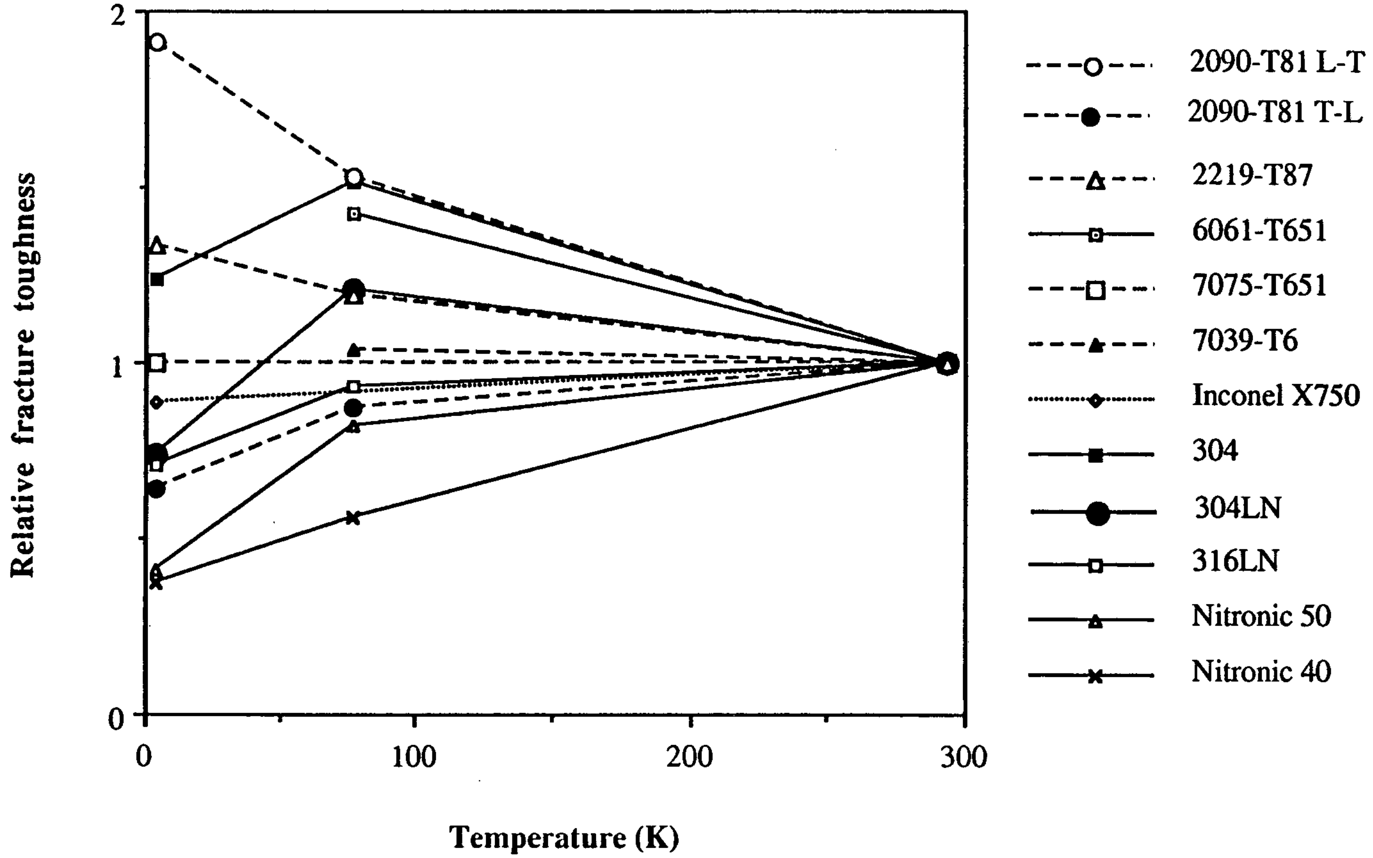
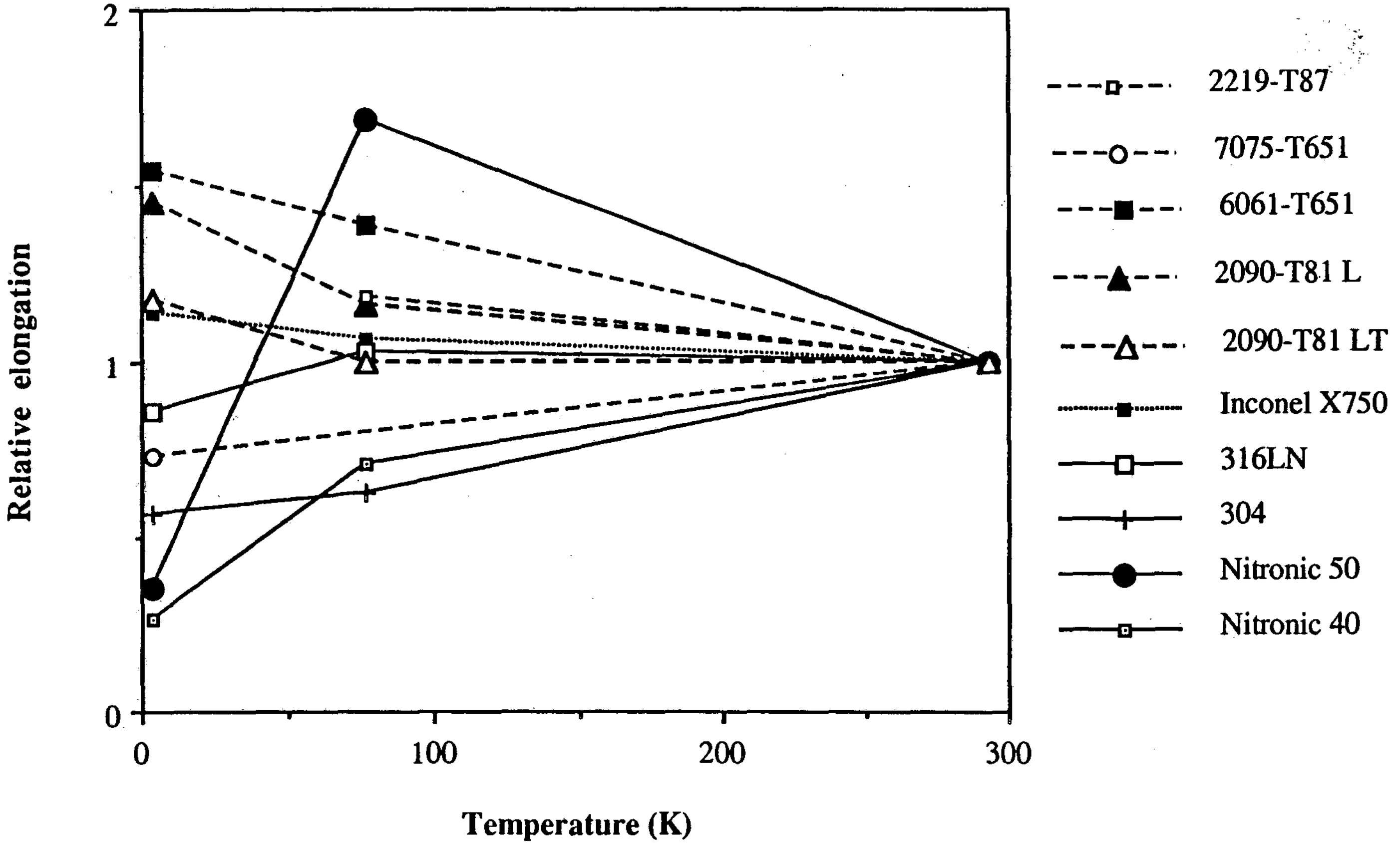
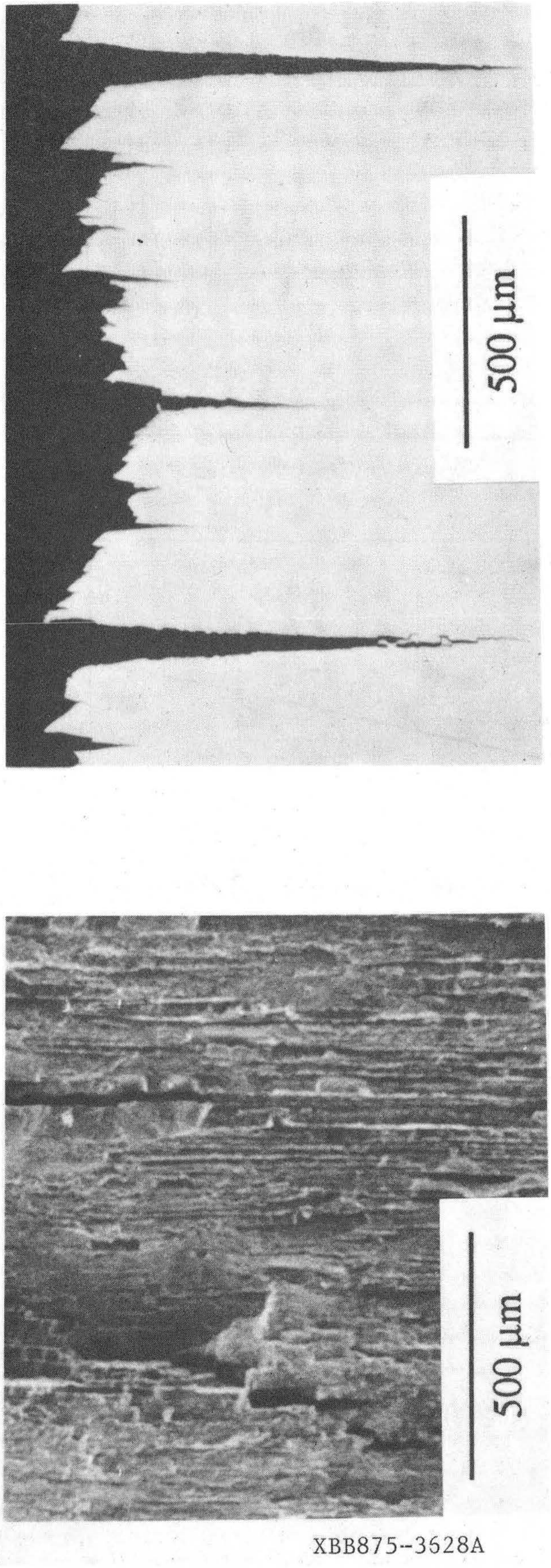

2
1
0
Relative fracture toughness
0
100
200
300
Temperature (K)
2090-T81 L-T
2090-T81 T-L
2219-T87
6061-T651
7075-T651
7039-T6
Inconel X750
304
304LN
316LN
Nitronic 50
Nitronic 40

Figure 3



2
1
0
Relative elongation
0
100
200
300
Temperature (K)
2219-T87
7075-T651
6061-T651
2090-T81 L
2090-T81 LT
Inconel X750
316LN
304
Nitronic 50
Nitronic 40

Figure 4



XBB875-3628A

Figure 5

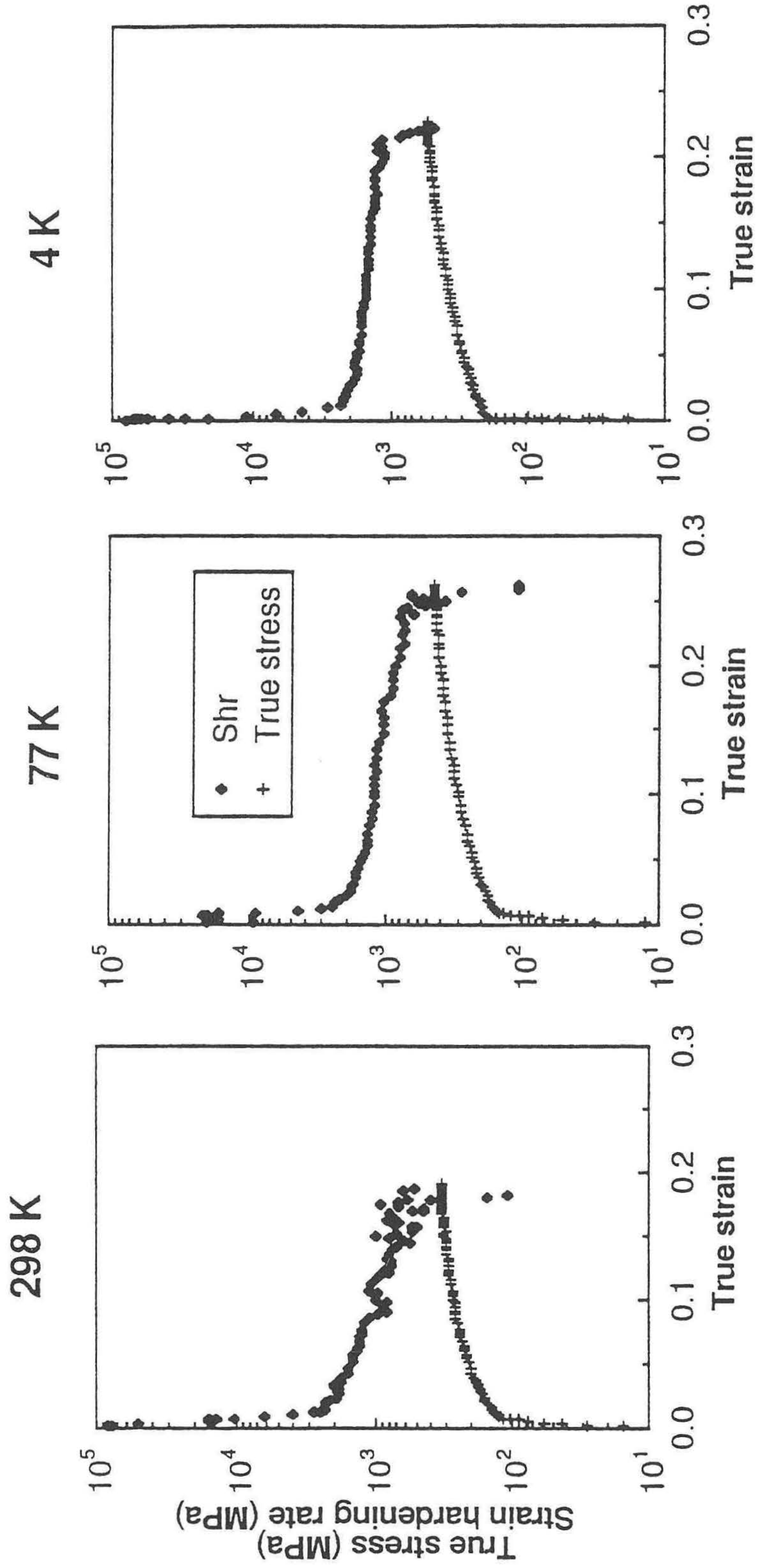

298 K
77 K
4 K
Strain hardening rate (MPa)
True stress (MPa)
10^5
10^4
10^3
10^2
10^1
0.0
0.1
0.2
0.3
True strain
◆ Shr
+ True stress

Figure 6

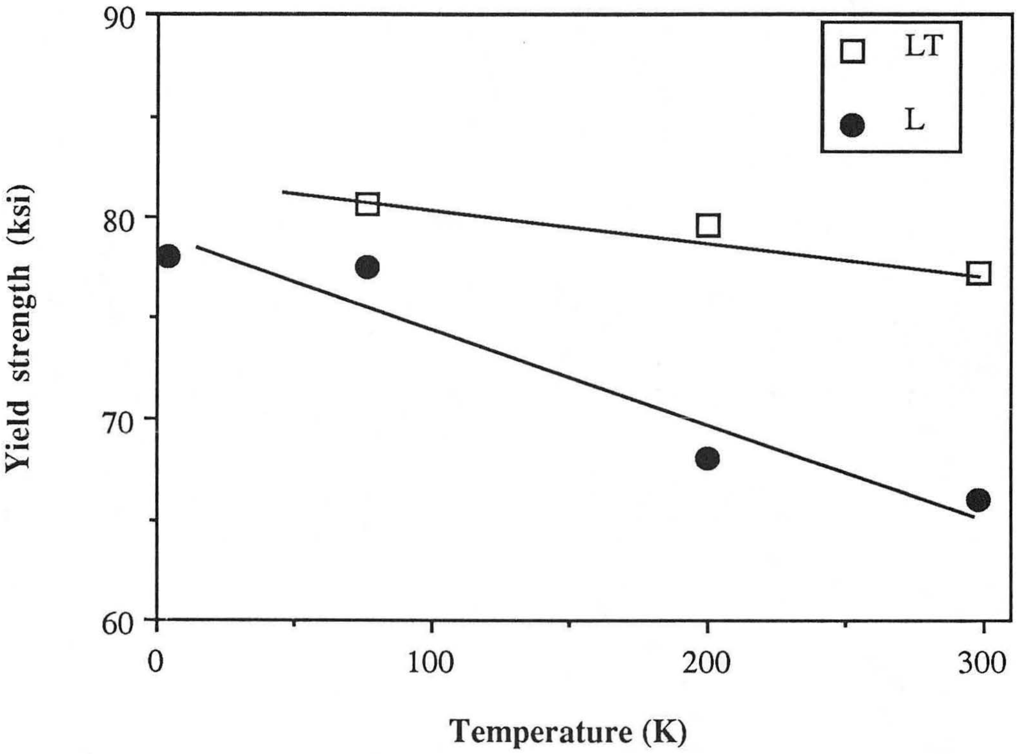


Figure 7