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PLASTIC FLOW AND STRENGTH OF URANIUM AND ITS ALLOYS

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ABSTRACT

The mechanical behavior of uranium and its alloys is reviewed with special emphasis on plastic flow behavior of polycrystalline materials. The strength of uranium is influenced by crystal structure as well as by point, planar, and volume defects; with these factors in mind, texture hardening, strain hardening, solute hardening, grain and subgrain size hardening, and precipitation and particle hardening are discussed. Temperature and strain rate influence the various strengthening mechanisms in different ways, and these variables are considered whenever possible. Unusual mechanical effects result from the polymorphic nature of uranium and from anisotropic properties of alpha uranium such as thermal expansion and elastic stiffness; their importance on plastic flow is considered. Uranium and its alloys can be made superplastic, and the factors which contribute to such characteristics are discussed.

I. INTRODUCTION

Plastic flow and strength characteristics of many metal-base materials are well documented, and much theoretical and phenomenological understanding has evolved in these areas in the past twenty-five years. Our interpretation of plastic flow and strength is based on an understanding of the crystalline structure of matter, of defects and their interactions within crystals, and of the nature of the bonding forces between atoms.

Uranium and its alloys can be considered excellent structural materials since they are elastically stiff (comparable to steel) and can be made to be very strong (yield strengths of over 250 ksi are readily attainable) and ductile. They are particularly suitable when high-density materials are desirable, since uranium is among the most dense of all elements, at a specific gravity of 19.07 (steel is about 7.85). Most of the mechanisms of hardening or ductilizing that are now well established for many materials can be directly applied to uranium. Examples are: texture hardening, strain hardening, solute hardening, grain and subgrain boundary hardening, and precipitation or particle (dispersion) hardening. This paper will center on these strengthening mechanisms as they relate to uranium, and we will consider their importance as a function of temperature. Uranium and its alloys can be made superplastic, and the factors contributing to such characteristics will be considered.

II. CRYSTAL STRUCTURE AND SOME PHYSICAL PROPERTIES OF URANIUM

Uranium has some special characteristics which are unique compared to many other structural metals. It is polymorphic and exists in three different crystalline forms: alpha (a structure which can be represented by two interpenetrating, base-centered orthohombic cells), beta (a complex tetragonal structure), and gamma (a body-centered-cubic structure). Alpha uranium exhibits a high degree of anisotropy with respect to its elastic and thermal expansion characteristics. Since these physical properties are important in influencing plastic properties, we indicate the nature of such anisotropy in the first two figures.

The influence of temperature on the elastic modulus of uranium is shown in Fig. 1. The solid curves refer to the modulus of nontextured polycrystalline uranium for the three different crystalline forms of the metal.(1) One notes a small discontinuity in the modulus of alpha uranium at very low temperature (~40K) where a second-order transformation to alpha prime occurs (apparently alpha prime has the same orthorhombic structure as alpha, except for a superstructure of magnetic domains).(2) The anisotropy of Young's modulus is shown for alpha uranium by the dashed lines. E_{max} is on the (100) plane, inclined approximately 37° to the [001] direction, and is about 10° away from the nearest neighbor direction. E_{min} is on a [010] direction up to 575K, and then in a direction 45° to the [001] direction in the (010) plane above 575K. Thus, we see that whereas the modulus of nontextured polycrystalline uranium is 29×10^6 psi at room temperature, it can be as high as 41.5×10^6 psi and as low as 21.4×10^6 psi for a textured sample. Therefore, when an external force is applied to a polycrystalline aggregate, each grain experiences a stress state that is dependent on its modulus in the direction of the stress, and this modulus is, in turn, a function of the grain's crystallographic orientation. If continuity at grain boundaries is to be maintained, the internal stress developed in some grains can be quite high due to the uneven distribution of load, leading to early plastic flow (microyield strengths in the order of 1000 psi have been noted).(3) At room temperature much of this plastic accommodation to distribute the loads more evenly is accomplished by twinning. The twinning of alpha uranium has been extensively studied.(4,5,6)

Another important observation to be noted in Fig. 1 is the large difference in modulus between alpha or beta and gamma uranium when compared at a common extrapolated temperature. Alpha and beta uranium are elastically stiffer than gamma uranium by a factor of three. This is a very big effect and one which has not been observed in other polymorphic elements.

These observations may help explain why many uranium alloys are quoted to have low moduli at room temperature.(7) The low values may arise from the presence of gamma phase in quenched uranium alloys, but of course they can also be explained in some cases by the presence of a texture in the alpha phase. Low modulus values may also be obtained by anisotropy of elastic properties, causing a low-load microyielding and the subsequent measure of a low tangent modulus from the stress-strain curve.

The linear thermal expansion coefficient is a strong function of crystallographic direction in alpha uranium,(8) as shown by the data given in Fig. 2. The data reveal that $\Delta\alpha = (\alpha_{[100]} - \alpha_{[010]})$ is not only large at low temperature, equal to about 20×10^{-6} per °C, but increases to 80×10^{-6} per °C at high temperatures. This very large anisotropy in the linear expansion coefficient leads to the generation of large internal stresses during thermal cycling of polycrystalline uranium. In turn, the internal stress leads to dimensional change of the sample, especially when under the influence of a small applied stress.(9) Thermal cycling can also lead to superplastic behavior in uranium,(10) and this aspect will be covered in a later section of the paper.

Atomic mobility is an important factor in determining the strength of crystalline solids at elevated temperature. For this reason we plot the self-diffusion coefficient of uranium(11-14) as a function of temperature in Fig. 3. As can be seen, the diffusivity of gamma uranium is considerably greater than those observed for alpha and beta uranium, which can be partly attributed to the open structure of body-centered-cubic gamma uranium. The diffusion coefficient for γ-U, however, is more rapid than predicted from the behavior of most other body-centered-cubic metals (compared at the same homologous temperature). For example, the diffusion coefficient at $0.75T_m$ is about 3×10^{-9} cm^2/sec for γ-U, whereas it is one order of magnitude lower for most b.c.c. metals.(15) Two sets of self-diffusion data are plotted in Fig. 3 for β-U. They differ by almost a factor of ten. Although no explanation has been given for this discrepancy, it is possible that it is due to a texture effect, β-U having a tetragonal structure where anisotropic diffusion can be expected.

III. STRENGTH AND DUCTILITY OF COMMERCIALLY PURE POLYCRYSTALLINE URANIUM

The effect of temperature on the tensile properties and ductility of commercially pure polycrystalline uranium (grain size about 0.05 mm) is summarized in Fig. 4. Tests were performed at a strain rate of about 10^{-4} sec^{-1}. At such strain rates, it is fairly well established that low-temperature mechanical behavior of polycrystalline solids is quite different from high-temperature behavior.(17) The temperature of demarcation is best described in terms of a homologous temperature. Low or cold temperature behavior is generally considered to be below $0.35T_m$. Warm temperature refers to deformation in the temperature range 0.35 to $0.6T_m$ and is characterized by considerable atom mobility (especially by dislocation short-circuiting), leading to formation of fine subgrains. High or hot temperature refers to deformation in the range above $0.6T_m$, where lattice diffusion dominates the deformation process and strain hardening is of limited importance. We will make use of this type of temperature differentiation in discussing the mechanical behavior of uranium and its alloys in this and subsequent sections.

A. LOW-TEMPERATURE BEHAVIOR ($<0.35T_m$)

Many interesting features can be noted relative to the low temperature mechanical behavior of alpha uranium (Fig. 4). A ductile-brittle transition temperature (D.B.T.T.) is observed at about 0°C. Fracture below the D.B.T.T. apparently occurs by cleavage at twin boundaries or by grain boundary separation.(3,18) The tensile strength is seen to decrease with a decrease in temperature in this range. This can possibly be explained by the increased importance of internal stress generated by elastic deformation (from elastic anisotropy) as the number of active slip systems diminishes with decreasing temperature. Although the tensile ductility of alpha uranium is low at low temperature, it can be deformed to large strains in compression. Deformation to strains in excess of 50% (true strain of 0.69), with no evidence of microcracking, has been obtained at temperatures at and below -150°C.(19)

The fatigue strength of alpha uranium has been studied in the temperature range 150°C to 600°C by Pedersen and Murphy.(20) We plot the endurance limit (stress for 5×10^8 cycles to failure) as a function of the corresponding ultimate tensile strength (U.T.S.) in Fig. 5. For many metals and alloys the ratio of endurance limit to U.T.S. is 0.5(22,23); this is shown for iron and titanium alloys in the same figure. The ratio for alpha uranium, however, is 0.15, indicating that the fatigue strength is much lower than expected from normal metal behavior. We would again like to postulate that this might be due to the ready generation of internal stress from elastic anisotropy during cylic stressing coupled with the limited slip systems available in uranium. It would be of interest to determine if gamma-stabilized uranium alloys (where many slip systems are expected) might behave in a more normal manner with respect to fatigue strength.

B. INTERMEDIATE-TEMPERATURE BEHAVIOR ($0.35 - 0.6T_m$)

In addition to the presence of a ductile-brittle transition temperature in alpha uranium, there is also a ductility minimum at about 300°C (Fig. 4). With the ductility minimum one observes an inflection or a maximum in the yield, tensile, and fracture strengths. The homologous temperature for these inflections is at about $0.4T_m$, a temperature where one might expect interaction of moving dislocations with moving Cottrell atmospheres of substitutional solute atoms.(24) In the case of commercially pure uranium, the impurity atoms would total about 300 to 500 atom parts per million (major impurities are Fe, Si, C, N, Ni, Mg, Mn). This is probably a sufficient amount to cause the anomalous strengthening effect observed. In aluminum-magnesium alloys, the maximum interaction of moving solute atoms with moving dislocations occurs at $\frac{\dot{\varepsilon}}{D_s} = 10^{15}\ \mathrm{cm}^{-2}$, where $\dot{\varepsilon}$ is the strain rate and D_s is the diffusion coefficient of substitutional magnesium atoms (generally about the same value as for matrix atom diffusion). We can make a similar calculation for the value of $\dot{\varepsilon}/D_s$ for alpha uranium. The self-diffusion coefficient of alpha uranium is given by $2 \times 10^{-3} \exp \frac{-40{,}000}{RT}\ \mathrm{cm}^2/\mathrm{sec}$.(11) Thus at 300°C, where the maximum in strength is observed (Fig. 4), $D_s \approx D_\alpha \approx 10^{-18}$ cm^2/sec. Since $\dot{\varepsilon} \approx 10^{-4}$ secs^{-1}, the value of $\frac{\dot{\varepsilon}}{D_s}$ is about 10^{14} cm^{-2}, which is in close agreement to that observed in the Al-Mg system. If this interpretation of the inflection in strength is correct, then we would predict that it will shift to higher temperatures as the strain rate is increased. One can solve for the temperature where this should occur by altering the relation $\frac{\dot{\varepsilon}}{D_s} = 10^{14}$ cm^{-2} as follows:

$$D_s = \frac{\dot{\varepsilon}}{10^{14}} , \tag{1}$$

$$D_s \approx 2 \times 10^{-3} \exp \frac{-40,000}{RT} \approx \frac{\dot{\varepsilon}}{10^{14}} , \tag{2}$$

or, rearranging and solving for T, we have

$$T = \frac{8741}{11.3 - \log_{10}\dot{\varepsilon}} . \tag{3}$$

Thus, for a strain rate of 1 per sec, the inflection temperature should be equal to $\frac{8741}{11.3} = 773$K = 500°C. In close agreement with our prediction, Buhler and Wagener$^{(25)}$ observed an inflection in the flow stress-temperature curve (from compression tests) at 500°C for strain rates in the order of 1 per second.

Other suggestions have been presented to explain the inflection in the strength/ductility-temperature curve for alpha uranium. These are (') a balance between the Peierls stress and thermally activated slip processes,(3) (2) intergranular cavitation through grain boundary sliding,(3) (3) a balance between twinning and slip,(20) and (4) the disappearance of twinning.(26) If someone has a source of very high-purity uranium, it might be worthwhile to determine if the inflection disappears.

Deformation at warm temperature is generally accompanied by extensive formation of subgrains.(27-29) Subgrain boundaries are difficult to detect by ordinary polishing and etching techniques, and they usually require transmission electron microscopy methods for their characterization. However, subgrains have recently been reported for uranium after warm working.(30,31)

C. HIGH-TEMPERATURE BEHAVIOR (>0.6T_m)

The high-temperature mechanical behavior of pure metals and solid solution alloys is well characterized and reliable quantitative predictions are possible in this range. Little strain hardening is exhibited in this temperature range, and materials are characterized by the steady-state flow stress at a given strain rate (or the steady-state creep rate for a given applied creep stress). It has been shown(27) that the flow stress is primarily a function of the strain rate, modulus, and the diffusion coefficient, given as follows*:

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*The constant 4×10^{-6} is based on the units used for $\dot{\varepsilon}$ and D. In this case and in subsequent equations used in this paper, $\dot{\varepsilon}$ is in sec^{-1} and D is in cm^2/sec.

$$\sigma = (4 \times 10^{-6})\ E \left[\frac{\dot{\varepsilon}}{D}\right]^{1/5} . \qquad (4)$$

The diffusion coefficient important in plastic flow is usually the lattice self-diffusion coefficient, D_L, at temperatures above $0.6T_m$. Below this temperature the diffusion coefficient along dislocations, D_p, together with lattice diffusion, appear to be important, and an effective diffusion coefficient has been used.(32) In this way, strength data at intermediate and high temperature can be correlated with Eq. (4). There is evidence that dislocation pipe diffusion controls the deformation process at intermediate temperatures for several pure metals(17,32) including uranium.(33)

The data above $0.6T_m$, shown in Fig. 4, can be considered as high-temperature data, and the relative strengths of alpha, beta, and gamma can be determined from the relation given by Eq. (4). Specifically, this equation predicts that the flow stress should be a unique function of the modulus if comparisons are made at the same value of the diffusion-compensated strain rate, $\dot{\varepsilon}/D$. The data in Fig. 6 reveal that α and γ uranium fall in line with this prediction in harmony with most other metals investigated.* The strength of β-U is greater than that predicted by Eq. (4). Two points are plotted for this element (Fig. 6) because there are two widely differing diffusivities reported for β-U (Fig. 3). It is not clear why uranium in the β phase is so

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*The flow stress values for the various elements given in Fig. 6 deviate from the line given because other variables influence high-temperature strength, principally subgrain size(34) and stacking fault energy.(35)

relatively strong. There are some unusual aspects to its high-temperature behavior which should be mentioned. One is that the measured activation energy for creep (Q_c = 72 kcal/mole) is considerably higher than that for lattice self-diffusion (Q_L = 43 kcal/mole). The other is that the material strain softens appreciably,(38) suggestive of texture changes during plastic flow. Strain softening in zinc has been attributed to the breakup of a texture which was developed during extrusion of the material previous to mechanical testing.(39) Further creep studies on beta uranium appear warranted.

IV. STRENGTHENING MECHANISMS

There are many known methods of strengthening crystalline materials. Systematic studies have been performed in model systems to determine the influence of change in structure on the corresponding change in mechanical properties. Investigation of uranium and its alloys seems to have progressed in a less systematic manner, often yielding a hodge-podge of results difficult to decipher and evaluate. We will attempt to cover some of the contemporary methods used to strengthen crystalline solids, especially as related to uranium and its alloys.

A. TEXTURE STRENGTHENING

We have already indicated the strong anisotropy of elastic properties in alpha uranium (Fig. 1). These results indicate that by development of a severe texture in uranium, one may be able to develop an elastically hard material with a directional modulus as high as 41 × 10^6 psi or an elastically soft material with a directional modulus as low as 21 × 10^6 psi.

Extensive and thorough studies have been performed by Lacombe and his colleagues(5,40) on the slip and twinning modes present in alpha uranium. Furthermore, these investigators determined the influence of temperature on the critical resolved shear stress (C.R.S.S.) for plastic flow in the active slip systems. This is shown by the data of Daniel et al.(5,40) in Fig. 7, where the C.R.S.S., τ, is divided by the shear modulus for the specific slip plane considered. A number of important points can be noted: (1) at any given temperature there is a wide variation of strengths for the various slip modes in alpha uranium; (2) each slip mode exhibits a low-temperature region where the stress increases rapidly with decrease in temperature; and (3) at high temperature, there appears to be an athermal region for plastic flow for three of the slip modes, namely (010) [100], (110) [1$\bar{1}$0], and (021) [1$\bar{1}$2].

The presence of only one easy slip mode at temperatures below about 300K would help to explain the low tensile ductility observed in alpha uranium at low temperatures (Section IIIA). Anisotropic elasticity theory has been utilized to show why the (010) [100] slip system may be the most prevalent one at room temperature.(42)

The wide variation in strengths for the various slip modes at a given temperature would suggest that strongly textured uranium should result in highly anisotropic plastic properties. The contribution of twinning to the deformation process, however, must also be considered, and therefore the precise strength of specifically textured uranium may be extremely difficult to predict. The data of Hughes et al.(30) on uranium, of Werkema(43) on U-Ti alloy, and of Bly(44) on a gamma-stabilized material all suggest that highly textured structures are probably not as common in fabricated uranium alloys as one might expect.

The high-temperature strength of alpha uranium may also be altered by texture hardening. We note, from Fig. 7, the large relative difference in C.R.S.S. for the four slip modes at 900K. Furthermore, the modulus anisotropy is very large at 900K, $\frac{E_{max}}{E_{min}} = 2.7$ (Fig. 1), and since modulus is important in controlling the high-temperature flow stress [Eq. (4)], we may expect some promising creep-resistant properties for textured alpha uranium.

Peix and Cizeron$^{(45)}$ developed various degrees of [100] texture in α-U by extruding uranium powders at various temperatures and extrusion ratios. They revealed that as the amount of [100] texture was increased, the tendency for growth of uranium rods under thermal cycling (20 ⇄ 550°C) decreased.

B. STRAIN HARDENING

Strain hardening is a potent method of strengthening materials. It is the primary method of increasing the strength of pure metals and solid solution alloys. Cold working generally leads to two major changes in the internal structure of polycrystalline metals: (1) the average dislocation density increases and (2) cells consisting of dislocation arrays and tangles form whose size diminishes as strain hardening proceeds.

The yield strength of a cold-worked material can be fairly accurately predicted from the Taylor equation, namely

$$\tau_y = \tau_o + \alpha\, Gb\sqrt{\rho}\ . \tag{5}$$

Here τ_y is the yield stress in shear, τ_o is resistance to dislocation flow from variables other than the presence of dislocations (e.g., the lattice friction stress, grain and cell boundaries, etc.), α is a numerical constant equal to about 0.5, G is the shear modulus, b is the Burgers' vector of the dislocation, and ρ is the dislocation density. If we let $\tau = \sigma/\sqrt{3}$ (Von Mises' relation) and $G \simeq 0.4\ E$, we can rewrite Eq. (5) for the tensile yield strength of a polycrystal as

$$\sigma_y = \sigma_o + 0.35\ Eb\ \sqrt{\rho}\ . \qquad (6)$$

We can estimate the yield strength of a cold-worked sample of alpha uranium at room temperature by means of Eq. (6). The dislocation density for a severely cold-worked material is about 10^{12} cm^{-2}. Letting $\sigma_0 = 40{,}000$ psi (the yield strength of annealed uranium), $E = 29 \times 10^6$ psi, and $b = 3 \times 10^{-8}$ cm, we obtained a yield stress σ_y of 350,000 psi. This is an impressive strength. Although no such values have been reported for cold-worked uranium, the high strain hardening rate for alpha uranium at room temperature, shown in Fig. 8, would imply that high strengths can be achieved. As can be seen, at a true strain of 0.6, the flow stress of uranium is about 175,000 psi. Investigations by Hockett et al.(46) revealed that flow stresses exceeding 250,000 psi are achievable after large compression deformation ($\varepsilon \simeq 3.0$).

Figure 8 also illustrates the difference in behavior of pure polycrystalline iron and alpha uranium. It would appear the α-U strain hardens considerably more rapidly than α-Fe. Both metals have about the same modulus, and their melting temperatures are not vastly dissimilar, so that room temperature is at about the same homologous temperature ($T \simeq 0.2T_m$).

An understanding of the nature of strain hardening in uranium has not been developed. Many investigators determine the strain hardening exponent, n, through the relation

$$\sigma = K\varepsilon^{n}, \tag{7}$$

where σ is the true flow stress, ε is the true plastic strain, and K is a material constant. The exponent n for uranium has been reported as equal to about 0.3 to 0.4 at room temperature, decreasing about linearly with temperature to a value of about 0.1 at 500°C, and remaining at this value beyond 500°C(16,47). These values were obtained in the region of strain where strain hardening is occurring. As can be seen, the stress-strain curve reaches a plateau at large strains (Fig. 8), yielding a steady-state flow stress, and here n would equal zero. At high temperatures, the steady-state flow stress is achieved at small strains, on the order of $\varepsilon = 0.1$ or less.

In addition to strain hardening from dislocation multiplication, the flow stress of several pure metals has been shown to be a function of the cell structure generated by plastic work (presumably a dynamic recovery process involving dislocation rearrangement into low-energy metastable arrays). Langford and Cohen(48) have shown that the yield strength of iron can be related to the cell size by the relation

$$\sigma = \sigma_0 + K'\lambda^{-1} , \tag{8}$$

where σ_0 is the stress of polycrystalline uranium without cells, λ is the cell size, and K' is a material constant related to the strength of the cell as a barrier.(49)

Neither Eq. (5) nor Eq. (8) has been applied to studies of the strength of alpha uranium as influenced by strain hardening. Such investigations should be performed, especially since very high strengths seem to be achievable in alpha uranium.

C. SOLID-SOLUTION HARDENING

Solid-solution hardening is a potent method of hardening crystalline materials, both at low and at high temperature. Examples that come to mind are nickel, zinc, and tin additions to copper (monel, brass, and bronze, respectively). We will consider the potential methods of strengthening uranium by solid solution alloying at low, intermediate, and high temperatures.

(1) Low Temperatures ($<0.35T_m$)

The mechanism of solid-solution hardening is not well understood, although empirically (guided by theoretical arguments) it has been shown that solute strengthening at low temperature can be correlated with size, valence, and modulus differences between solute and solvent atoms. Solute strengthening has been extensively studied in copper and its alloys and will be described as a model system in the following discussion. Early work suggested that size difference was the major factor that lead to strengthening(50-52); the greater the absolute difference in size of solvent and solute atoms the greater the increase in strength. This strengthening is currently attributed to Cottrell locking, that is, the interaction of solute atoms with the hydrostatic stress fields of edge dislocations.(24) An example for such a size effect for solid-solution alloys of copper is shown in Fig. 9A. Although the overall correlation is quite good, deviations with certain solute elements are evident. Studies by Dorn and his colleagues on aluminum solid solution alloys(53) and Hibbard on copper solid solution alloys(54) suggest that valence (or electronic) differences between solute and solvent are important. In fact, Hibbard suggested that the size effect difference of Fig 9A can be principally accounted for by a valence difference. The rather good correlation shown in Fig. 9B with certain solutes would suggest that Hibbard's ideas may have some merit. It should be noted, however, that certain elements (Sn, Sb, In, Ag, Au) lead to higher hardening than predicted by Hibbard. Theoretical justification for the valence or electronic effect is usually centered on the contribution of Suzuki locking(55) to strengthening; that is, the interaction of solute atoms with the stacking fault region of partial dislocations. More recently, Fleischer(56) concluded that the inherent modulus of solute atoms was an important factor in contributing to solid solution strengthening. His calculations, guided by experimental data, led him to conclude that modulus and size differences could account for solid-solution strengthening in copper alloys. The correlation obtained with Fleischer's parameter is shown in Fig. 9C. Hutchison and Pascoe(57) concluded that none of the parameters proposed can fully explain the experimental data for copper solid-solution alloys. They proposed a parameter which takes into account size differences and differences in relative compressibilities of the solute atoms, which leads to a better correlation than those shown in Fig. 9.

Similar studies for uranium, as those made for copper and aluminum alloys, have not been performed. Certain difficulties arise. For one, few elements dissolve readily in alpha uranium, and therefore little in meaningful research can be performed except at very dilute concentrations. On the other hand, many elements dissolve readily in bcc gamma uranium. These solid solutions, however, are usually metastable at low temperatures, and complex transformations often occur, sometimes as a result of strain during testing(58,59), making analysis of the resulting mechanical properties difficult to interpret correctly.(7,59,60,61) In addition, the electronic contribution to strength by solid solution alloying is difficult to analyze because of uncertainties in the valence state of uranium.(62)

(2) Intermediate and High Temperature ($>0.35T_m$)

Mention was already made of the possible contribution of solid solution alloying to strengthening at intermediate temperatures where interactions can occur between dislocations with moving Cottrell atmospheres (Section IIIB). It was suggested that such solute strengthening effects should occur at $\dot{\varepsilon} = 10^{15}\ \mathrm{cm}^{-2}\ D_s$, where D_s is the diffusion coefficient of solute atoms. Further studies are needed here to determine the magnitude of the effect in relation to the nature of solute atoms (e.g., size difference). In addition, the Portevin-Le Chatelier effect (serrated stress-strain curves) should be present if the postulate we have made is correct with respect to the interaction of moving solute atoms and moving dislocations. This observation has not been noted, although serrated stress-strain curves are sometimes obtained at lower temperatures(40,63) than predicted by Eq. (3).

Strengthening by solid solution alloying for elevated temperature service has been studied, but in a rather haphazard manner.(64) We are able, however, to predict the strengthening expected from the presence of solute atoms. If the alloying element added is not too dissimilar in size and modulus (Class II solid solution alloys)(65) the strengthening (or weakening) expected can be predicted from Eq. (4), namely

$$\sigma \approx 4 \times 10^{-6} E \left(\frac{\dot{\varepsilon}}{D_{alloy}}\right)^{1/5} , \qquad (9)$$

where D_{alloy} is the average diffusion coefficient of solute and solvent atoms. Thus, if the average modulus, E, is increased and D the diffusivity, D_{alloy}, is decreased by solute additions, solid solution strengthening will occur.

When the size and modulus of solute atoms are quite dissimilar from the host atoms (Class I solid solution alloys), then another equation describes the high-temperature strength behavior and is given as follows:

$$\sigma \approx 3 \times 10^{-6} E \left(\frac{\dot{\varepsilon}}{D_{alloy}}\right)^{1/3} . \qquad (10)$$

In this equation, D_{alloy} is a complex function of the individual diffusion coefficients of the solute and solvent atoms.(29)

Solid-solution hardening can be a powerful method of increasing the strength of uranium, especially in the gamma phase where foreign atoms can be made to dissolve readily. For example, Mulberry (uranium containing 7.5%Nb and 2.5%Zr) has thirty times the strength$^{(44)}$ of pure gamma uranium at 800°C (at $\dot{\epsilon} = 10^{-4}$ secs^{-1}). This arises primarily from the decrease in diffusion coefficient by alloying (about 10,000 fold decrease) with zirconium and niobium. Another factor is the interaction force between solute atoms and dislocations. The validity of Eq. (10) is shown in Fig. 10 for Mulberry creep tested in the gamma range where the slope of the log σ vs log $\dot{\epsilon}$ plot is shown to have a slope of about 1/3.

D. GRAIN AND SUBGRAIN BOUNDARY HARDENING

Grain boundaries are considered to be effective barriers to plastic flow at low temperatures. Most researchers consider that the Hall-Petch relation adequately describes the influence of grain diameter, d, on the strength of metals and alloys. This equation is given as

$$\sigma = \sigma_o + K_y\, d^{-1/2}, \qquad (11)$$

where σ is either the yield strength, flow stress at a given strain, or sometimes even the fracture stress, σ_o is the stress needed for a dislocation to overcome the lattice friction and the stress fields of other dislocations, and K_y is a material constant believed associated with the stress to unpin a dislocation from its source. Several investigators have applied the Hall-Petch relation to an analysis of the mechanical behavior of uranium.

Grain size refinement generally not only leads to strengthening but also to ductilizing. An example of such a trend for uranium at room temperature is shown in Fig. 11. As can be seen, both the fracture stress, σ_F, and the percentage of elongation to fracture is increased with a decrease in grain size. The relation between σ_F and $d^{-1/2}$ is not linear, however, as predicted by the Hall-Petch relation. Taplin discusses(66) in detail the inadequacies of the Hall-Petch relation for describing the strength of uranium; he considers the complications rising from determining a true yield strength in uranium and from the fracture process itself as influenced by twinning (which, in turn, is influenced by grain size) and concludes that no simple correlation between σ and d should be expected. In spite of these difficulties it is rather clear that grain size refinement is a powerful method of decreasing the ductile-brittle transition temperature and in increasing the strength of uranium.

Subgrain boundaries should also be effective in increasing the strength of uranium and in inhibiting fracture. Well-defined subgrains are usually developed by warm working ($0.35 - 0.6T_m$), where sufficient recovery can occur for dynamic polygonization. Bolton and Tilbury(67) have shown that warm working can reduce the ductile-brittle transition temperature substantially. Figure 12 illustrates the ductility characteristics of uranium as influenced by warm working at 250°C ($T = 0.43T_m$, where $T_m = 1225K$, the theoretical melting temperature for alpha uranium).(68) It can be seen that by increasing the warm-rolling reduction from 10 to 40%, the transition temperature is lowered by about 60°C. More recently, Tilbury(69) showed that a combination of vacuum annealing and warm working appears to lower the transition temperature more than either separately. These results suggest that gaseous impurities strongly influence mechanical properties. It is well established that hydrogen in small quantities is very detrimental to the ductility of uranium.(70,71) Tilbury and Bolton could find no evidence for subgrains in their warm-worked sheet by optical metallography and x-ray back reflection techniques. A more definitive approach for detecting subgrain boundaries, however, is by transmission electron microscopy (TEM). Subgrains in alpha uranium have been observed by Guillaume and Lions after warm temperature annealing of cold-worked uranium using TEM techniques.(72)

Hughes et al.(30) have studied the mechanical anisotropy of warm rolled uranium plates and found it to be small. They quote their coworkers Smith and Ruckman,(31) who apparently have observed dislocation substructures with a cell size of 0.36 to 0.46 microns in plates reduced by rolling between 70 and 90% in thickness at 250°C. Thus it seems certain that subgrains do develop under warm working conditions for alpha uranium in the same way as observed for most metallic alloys. Studies with iron-base alloys have revealed that fine subgrains can lead to very high yield strengths following the relation given by Eq. (8). No direct correlations between subgrain size and properties of uranium and its alloys were revealed in our search of the literature, and studies on this technologically important subject should be initiated.

E. PRECIPITATION AND DISPERSION HARDENING

Precipitation hardening is very likely the most potent method of strengthening uranium and its alloys. A number of papers are being presented on this subject in the present symposium, and therefore our treatment of it here will be brief.

Equilibrium phase diagrams indicate that the hardening precipitate in quenched uranium alloys containing small binary additions of titanium or molybdenum should be the nearby intermetallic compound as a result of the reaction

$$\alpha'_a \rightarrow \alpha + \delta$$

where we are using the nomenclature of Lehmann and Hills.(73) Ammons and Zukas indicate that this is the case.(74,75) There are other binary alloys of uranium (with Re, Rh, Ru, Os, Pd, Ni, Co, Fe, and Mn) which have uranium-rich intermetallic compounds and which might be expected to exhibit precipitation hardening. The literature is not extensive on these systems, but apparently many of them are plagued with β phase retention on quenching.

It is evident, however, that precipitation hardening of uranium alloys is not always dependent upon a uranium-rich intermetallic compound. The uranium-niobium alloys, which have been extensively studied by several laboratories,(76,77) are probably the best such examples. There are no compounds in the U-Nb system.

More complex precipitation-hardening alloys containing several alloying elements have been investigated by Tardif,(78) Erickson,(79) and by Greenspan and Rizzitano.(80)

Dispersion hardening is not vastly dissimilar from precipitation hardening, since in both cases a second phase is the source of the strengthening process. Dispersion-hardened materials, however, do not generally have long-range stress fields associated with the particles (i.e., no high coherency stresses); in addition the particles are generally more stable than in precipitation-hardened systems, and they normally do not grow during use of the material.

Gurland(81) has summarized the influence of carbide particles on the strength of iron-carbon alloys at low temperatures and has suggested the relation

$$\sigma = \sigma' + K' D_s^{-1/2}, \tag{12}$$

where σ and K' are material constants and D_s is the interparticle spacing or the mean free ferrite path, depending on the volume fraction of cementite present. For large volume fractions of cementite, D_s is the interparticle spacing, whereas for low volume fractions D_s is the mean free ferrite path. Correlations of this type do not appear to have been made for dispersion-hardened uranium alloys.

Dispersion hardening can also be a very potent method of strengthening materials for high-temperature applications. The hardening that results from the presence of particles may be due mainly to the stabilization of the fine subgrain structure that is developed during the processing of the material. It has been suggested(28,32) that for low-particle-content, dispersion-hardened materials, the flow stress is given by

$$\sigma = \text{const. } E \left(\frac{\lambda}{b}\right)^{-2/7} \left(\frac{\dot{\varepsilon}}{D}\right)^{-1/7} . \tag{13}$$

Here. λ is the subgrain size and b is Burgers' vector. Bufferd, Zwilsky, and Grant$^{(82)}$ studied the creep properties of uranium containing alumina particles. They found that the material was considerably stronger than pure alpha uranium at 600°C (Fig. 13). The slope, m, of the log σ vs log $\dot{\varepsilon}$ curve is about 1/7 for the low-volume-fraction composite (3.5 w% Al_2O_3), as predicted by Eq. (13). The slope, however, is less than 1/7 for the high-volume-fraction alumina composite (7.5 wt% Al_2O_3). Furthermore, the activation energy for creep of this material is greater than that for lattice self-diffusion. Both observations are consistent with the behavior of large-volume-fraction particulate composites.$^{(83)}$

V. SUPERPLASTICITY OF URANIUM AND ITS ALLOYS

The term "superplasticity" refers to large neck-free elongations obtained during low-stress high-temperature deformation of certain metallic materials. Usually such materials are weak at temperatures where they are superplastic. There is a great deal of interest in such materials because they can be shaped easily. For example, many of the techniques used in forming plastics can also be used to form superplastic metallic base alloys.$^{(84-89)}$

Superplastic behavior seems to be associated with high strain-rate sensitivity. In the flow stress-strain rate relation

$$\sigma = K\dot{\varepsilon}^{m}, \tag{14}$$

m is known as the strain rate sensitivity exponent. Several authors$^{(89-92)}$ have shown that as the sensitivity exponent increases the rate of local deformation or necking decreases, leading to greater total elongation. Materials exhibiting superplasticity generally have m values ranging from 0.5 to 1.0 [as noted earlier, m is 0.2 for most pure metals, Eq. (4), and 0.33 for some solid solution alloys, Eq. (10]. Two distinctly different types of superplastic flow have been noted in metallic alloys: (a) internal-stress plasticity and (b) fine-structure plasticity. We will now discuss these especially as related to uranium and its alloys.

A. INTERNAL-STRESS PLASTICITY

Internal generation of stress during plastic flow can lead to high strain-rate sensitivity, high plasticity, and low strength.

One method of obtaining superplasticity of this type is to cause a continuous phase transformation under the simultaneous presence of an externally applied load. The volume change during phase transformation creates internal stress. Thus, polymorphic metals undergoing phase transformation should exhibit superplasticity. Studies on such metals have revealed that the strain produced from the phase transformation is proportional to the externally applied stress.$^{(93-95)}$ Furthermore, the plastic strain (at a given applied stress) induced from phase transformation increases with increasing difference in the volume change occurring during transformation and with decreasing strength of the phases involved.$^{(96,97)}$ In equation form, we can write

$$\varepsilon_{\alpha \rightleftarrows \beta} = \text{const} \ \frac{\Delta V}{\sigma_{\alpha,\beta}} \cdot \sigma \ , \tag{15}$$

where $\varepsilon_{\alpha\rightleftarrows\beta}$ is the strain from a two-way transformation, ΔV is the absolute change in volume from α to β, $\sigma_{\alpha,\beta}$ is the strength or hardness of the weakest of the two phases involved, and σ is the applied stress. The validity of Eq. (15) for a number of polymorphic metals is shown in Fig. 14. Since the rate of heating and cooling through the transformation is usually about the same in most investigations (about 1°C per minute), one can consider that the time during transformation, when the strain $\varepsilon_{\alpha\rightleftarrows\beta}$ occurs, is a constant. Thus, Fig. 14 also represents the relation between strain rate, $\dot{\varepsilon}_{\alpha\rightleftarrows\beta}$ and the applied stress, σ. Since the relation observed is about linear (at least at low stresses), m is equal to unity and superplasticity can be expected. Indeed, materials undergoing continuous phase transformations can be deformed large amounts, typically several hundred percent, before ultimate failure occurs.(95)

Another method of generating internal stress is by thermal cycling with polycrystalline materials that exhibit thermal expansion anisotropy. During temperature changes, each grain expands or contracts in a direction different from its neighboring crystals, which creates stresses at the boundary regions of each grain. Uranium exhibits very high thermal-expansion anisotropy; the data in Fig. 2 illustrate that $\Delta\alpha$ is large at room temperature and increases with an increase in temperature. The high internal stress generated under thermal cycling assists the plastic flow process leading to a high strain-rate sensitive material under such conditions.(100) An example for alpha uranium, illustrating this behavior, is shown in Fig. 15. In this figure the flow stress (compensated by modulus) is plotted as a function of flow rate (compensated by the diffusion coefficient*). As can be seen, the strength of the thermally cycled samples, σ_T, is lower than the strength of specimens deformed isothermally, σ_I; for example, at $\frac{\dot{\varepsilon}}{D} = 10^8\ \mathrm{cm}^{-2}$, $\sigma_T \simeq 0.5\ \sigma_I$. More importantly, the strain-rate sensitivity exponent is higher for the thermally cycled sample ($m = 1$ at low stresses) than for the isothermally tested sample ($m = 0.17$). Thus, superplastic flow is expected in thermally cycled samples, and this has been observed by Johnson and his colleagues.[10,97,100]

B. FINE STRUCTURE SUPERPLASTICITY

In dealing with superplasticity associated with fine structures, one is concerned with a structure that appears to remain the same during extensive plastic flow. Apparently internal stresses do not play an important role during deformation of such materials. An example of the fine structure present in a superplastic uranium alloy is shown by the photomicrographs in Fig. 16. This monotectoid composition alloy, known as Mulberry, consists of uranium, niobium (7.5 wt%), and zirconium (2.5 wt%). Appropriate thermal-mechanical processing results in the development of a fine, equiaxed, two-phase structure (α phase of nearly pure uranium plus solid solution γ phase). The resulting microstructure is shown in Fig. 16A. After 65% tensile deformation

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*It is necessary to use a diffusion coefficient which represents the average diffusivity during the variable temperature-time cycle; this was done by graphical integration using the relation $D_{avg} = \left[D_0 \int_0^{t_1} \exp\,(-Q_L/RT)\ dt \right] \Big/ t_1$, where t_1 is the time for a complete thermal cycle.

at 625°C, the structure remains equiaxed (Fig. 16B), although slight growth of the gamma phase has occurred. These photomicrographs indicate that one is not dealing with phase or volume changes, at least not with the large volume change that occurs during phase-transformation plasticity. There must be, however, a great deal of atom mobility taking place during superplastic flow in order to permit the interphase boundaries to migrate and maintain the same shaped grain structure as the external shape of the specimen changes drastically.

Structurally, the principal prerequisite for isothermal superplasticity in metallic alloys is the presence of a stable fine-grained structure. Generally, the grain size is of the order of 1 to 5 microns. Such alloys are usually superplastic in the temperature range 0.5 to 0.7 of their absolute melting temperatures. Above $0.7T_m$, the structure often coarsens, leading to loss of superplasticity (with uranium alloys phase transformation usually intervenes). Most superplastic alloys consist of two phases; the presence of the second phase apparently obstructs the growth of the fine grains in the matrix phase. A requirement of the second phase is that it should have about the same strength and ductility as the base alloy; apparently this permits it to deform with about the same ease as the base material.* It is

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*It should be emphasized that although a fine grain size appears to be a prerequisite for superplasticity in polycrystalline solids, it is not a sufficient condition. For example, if the grain boundaries are brittle (as in some ceramic polycrystals) or cannot migrate (as in dispersion-hardening materials) the fine-grained material can be quite brittle, even at high temperatures.

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VI. SUMMARY AND CONCLUSIONS

We have pointed out that uranium alloys are comparable to steels in their potential for mechanical properties, and that many of the fundamental mechanisms used for modification of properties of other metals are applicable to uranium alloys. On the other hand, we have emphasized the problems with the unusually large anisotropy of thermal expansion, and of modulus, in the crystals of alpha uranium. These lead to a nonuniform stress distribution in polycrystalline material — a form of stress concentration.

We considered the effect of strengthening mechanisms and strain rates in three different temperature regions. At low temperature we showed strong effects of anisotropy of properties on fatigue strength and on the tensile ductility. We briefly reviewed some of the strengthening mechanisms and suggested that subgrain size may be a potent factor in improving properties. In the intermediate temperature region, we suggested that interaction of Cottrell atmospheres with moving dislocations may play a role in determining the strength and ductility of commercially pure uranium and suggested the use of very high-purity uranium to test that idea. We discussed high-temperature properties as a function of strain rate, modulus, and diffusion coefficient as they appear in the equation

$$\sigma = (4 \times 10^{-6})\ E \left(\frac{\dot{\varepsilon}}{D}\right)^{1/5} . \qquad (4)$$

We noted the departure of beta uranium from this scheme and suggested more work on the creep of this material.

There are two types of superplasticity exhibited in uranium and its alloys which we discussed. The first type is due to internal stresses combined with low external loads. The internal stresses are produced by cycling through a phase transformation or by merely thermal cycling in the alpha region. The second type of superplasticity is due to an extremely fine grain size which is stabilized at an appropriate elevated temperature.

As evidenced by the recent literature and by many of the papers presented at this conference, considerable progress is being made toward understanding the problems associated with uranium and some of its simple alloys. Many of the phenomena and mechanisms observed in other metallic systems seem also to apply to uranium and its alloys. For examples we list hydrogen embrittlement, superplasticity, and several hardening mechanisms.

The future of uranium as a structural material for special applications seems quite promising, since it has the inherent strength and ductility necessary for such application, combined with its unusual physical properties.

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FIGURE CAPTIONS

Fig. 1. Young's modulus of pure polycrystalline uranium as a function of temperature. The modulus of nontextured uranium is given by the solid line. The maximum and minimum moduli for alpha uranium from single crystal measurements are also plotted.(1,2)

Fig. 2. Linear thermal expansion coefficient along the three principal crystallographic directions of alpha uranium. The influence of temperature on the anisotropy of thermal expansion is also illustrated. After Lloyd and Barrett.(8)

Fig. 3. Self-diffusion data for alpha, beta, and gamma uranium. Data from Refs. (11), (12), (13) and (14).

Fig. 4. Effect of temperature on the tensile properties and ductility of polycrystalline uranium. Tests performed at strain rates of about 10^{-4} $\sec^{-1}$.(3,16)

Fig. 5. Fatigue strength of alpha uranium(20,21) as a function of the ultimate tensile strength (U.T.S.) compared with other common metals.

Fig. 6. Relation between high temperature flow stress (at constant strain-rate/diffusivity ratio) and elastic modulus for a number of crystalline materials.(27,36,41)

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Fig. 9. Factors influencing solid solution strengthening of copper. (A) Flow stress variation with size difference between solvent and solute atoms. (B) Flow stress variation with valence difference between solvent and solute atoms. (C) Flow stress variation with size and modulus difference between solvent and solute atoms. Hutchison and Pascoe.(57)

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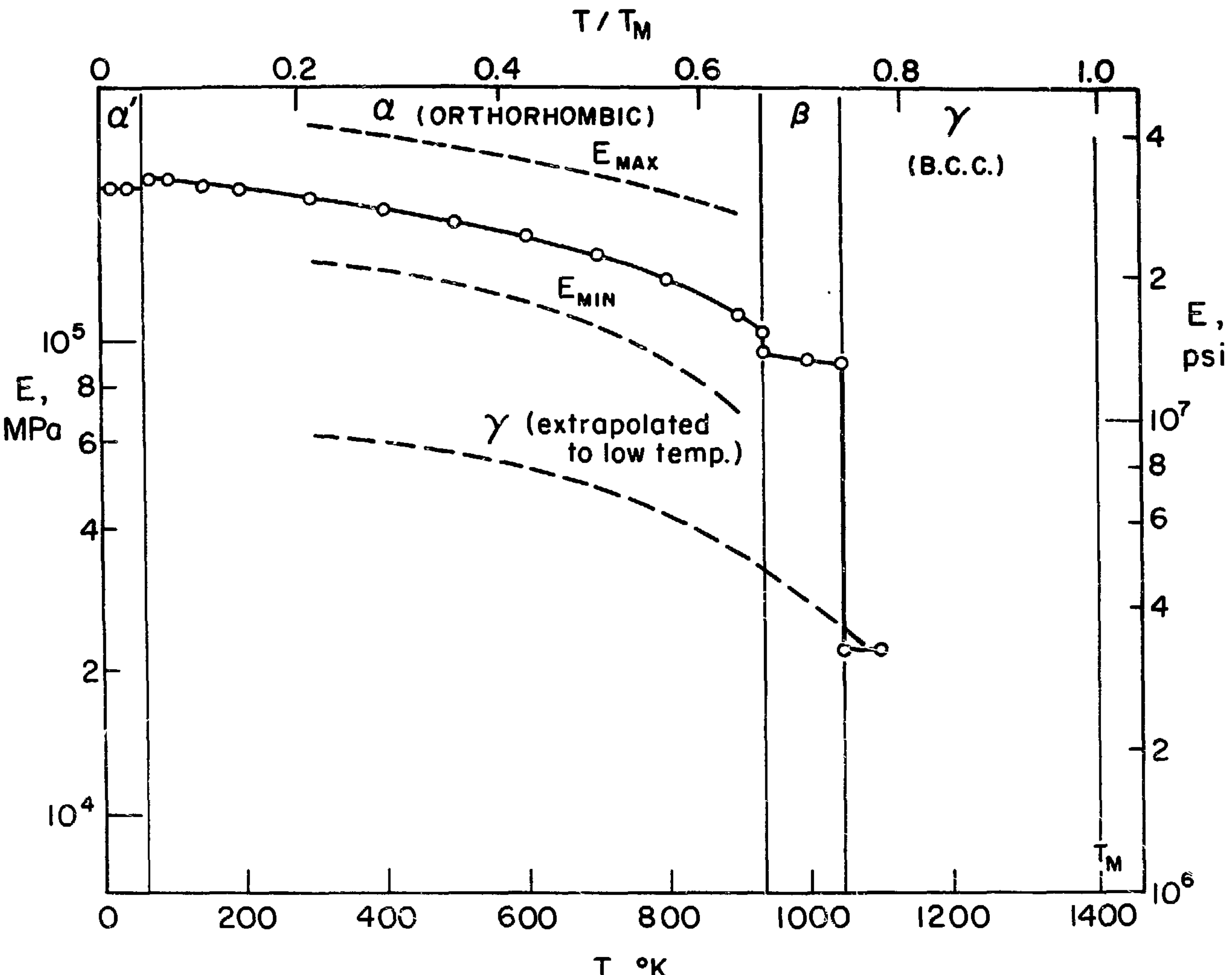

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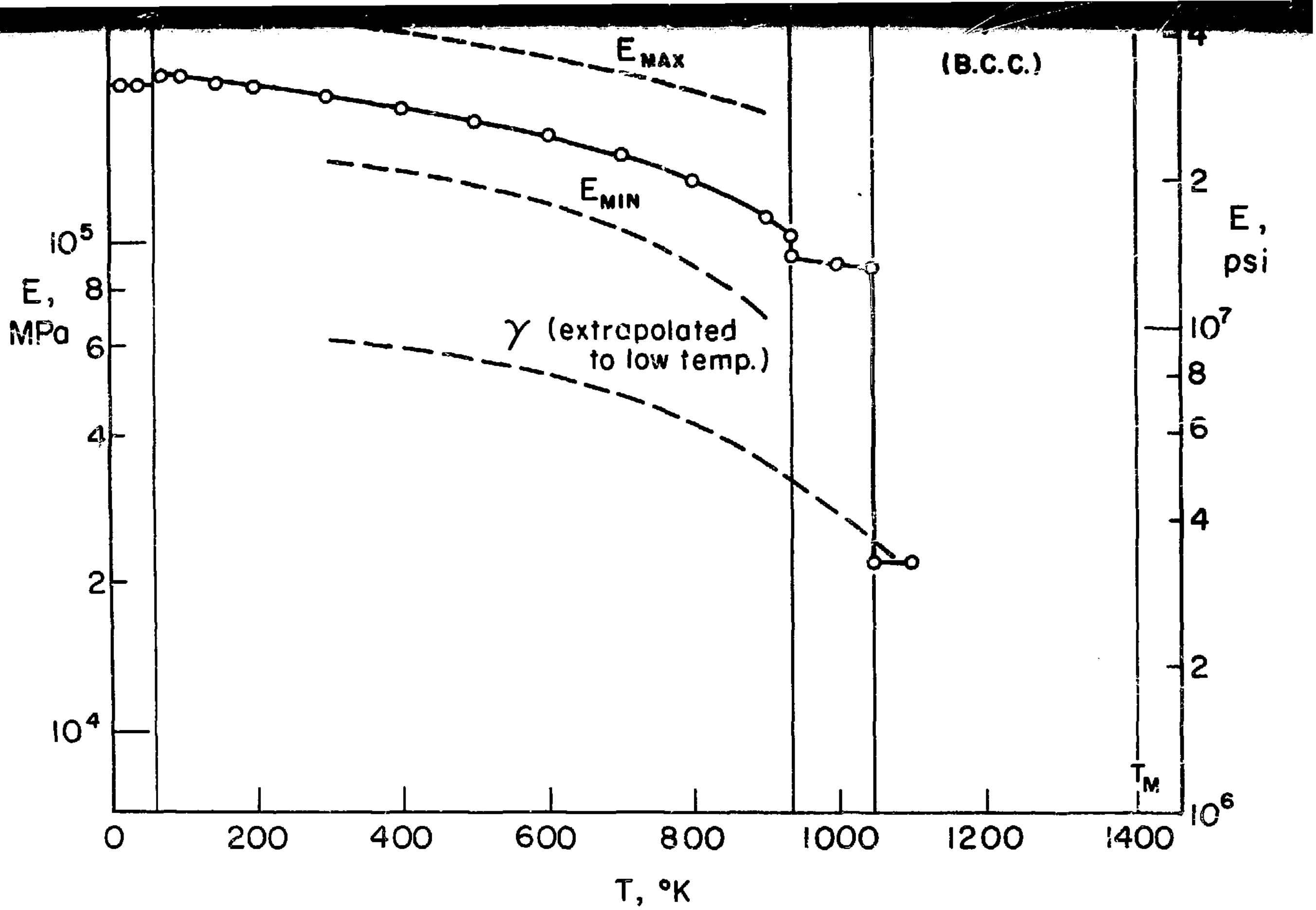

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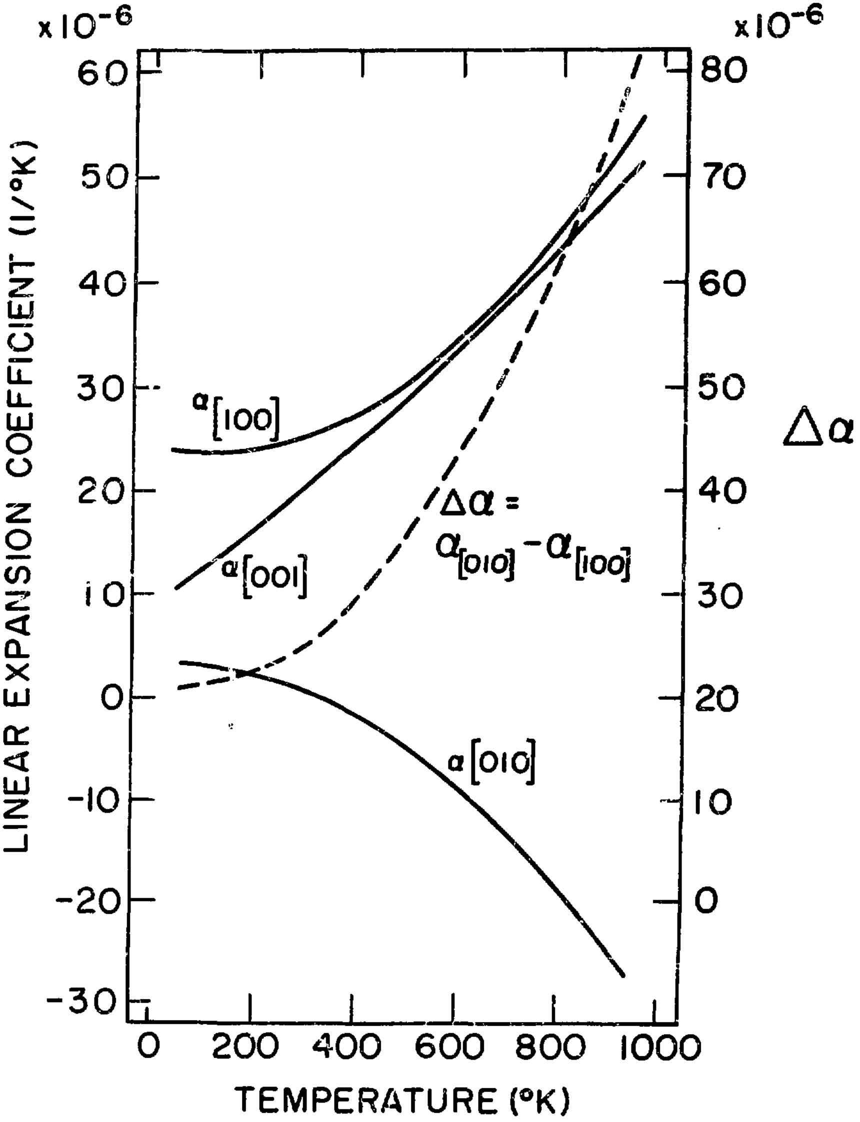

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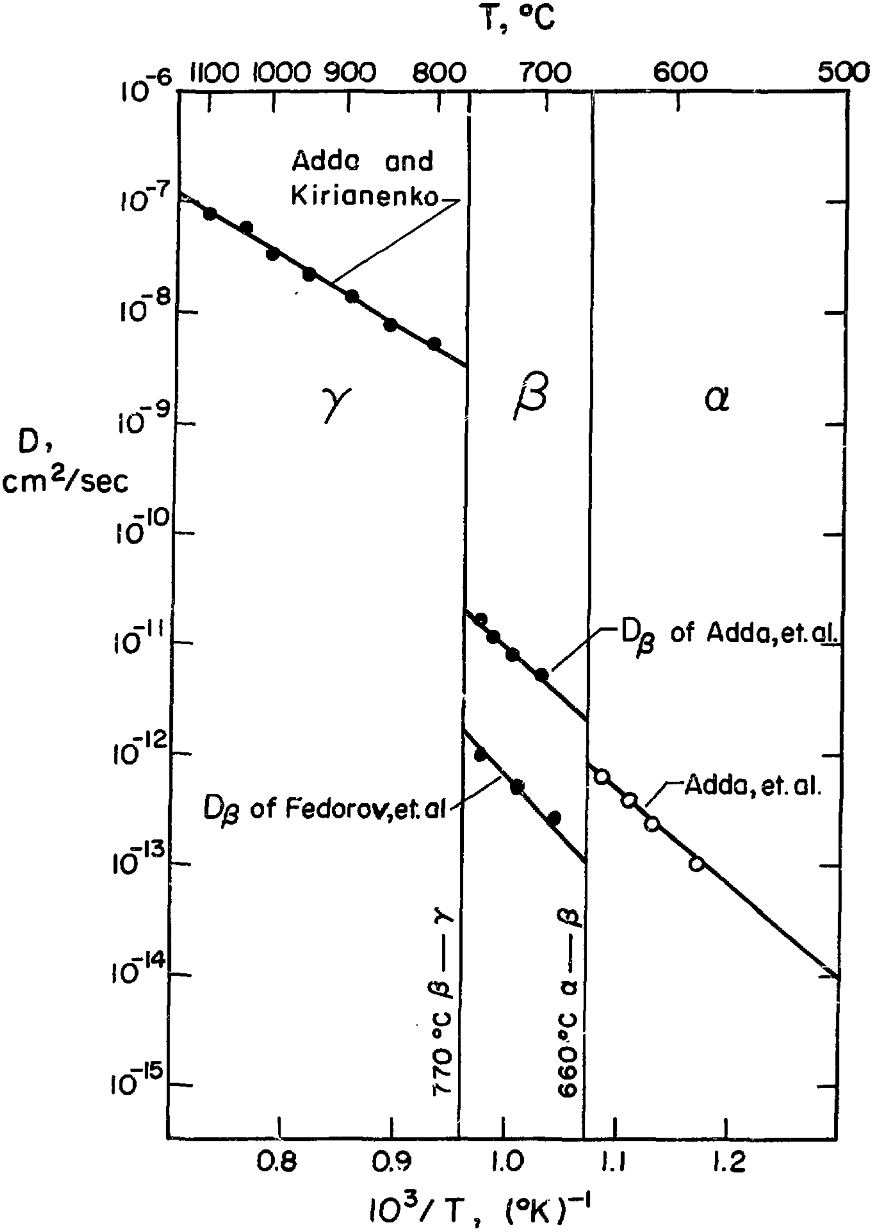

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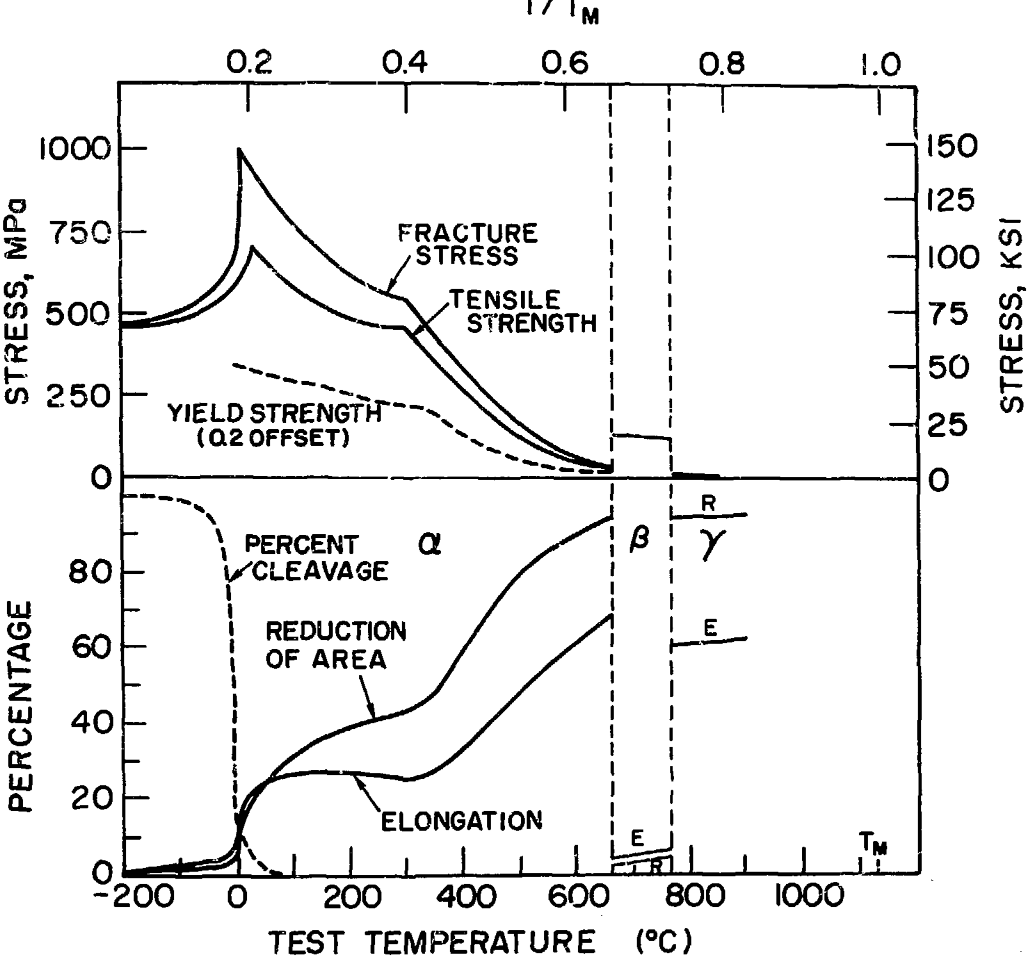

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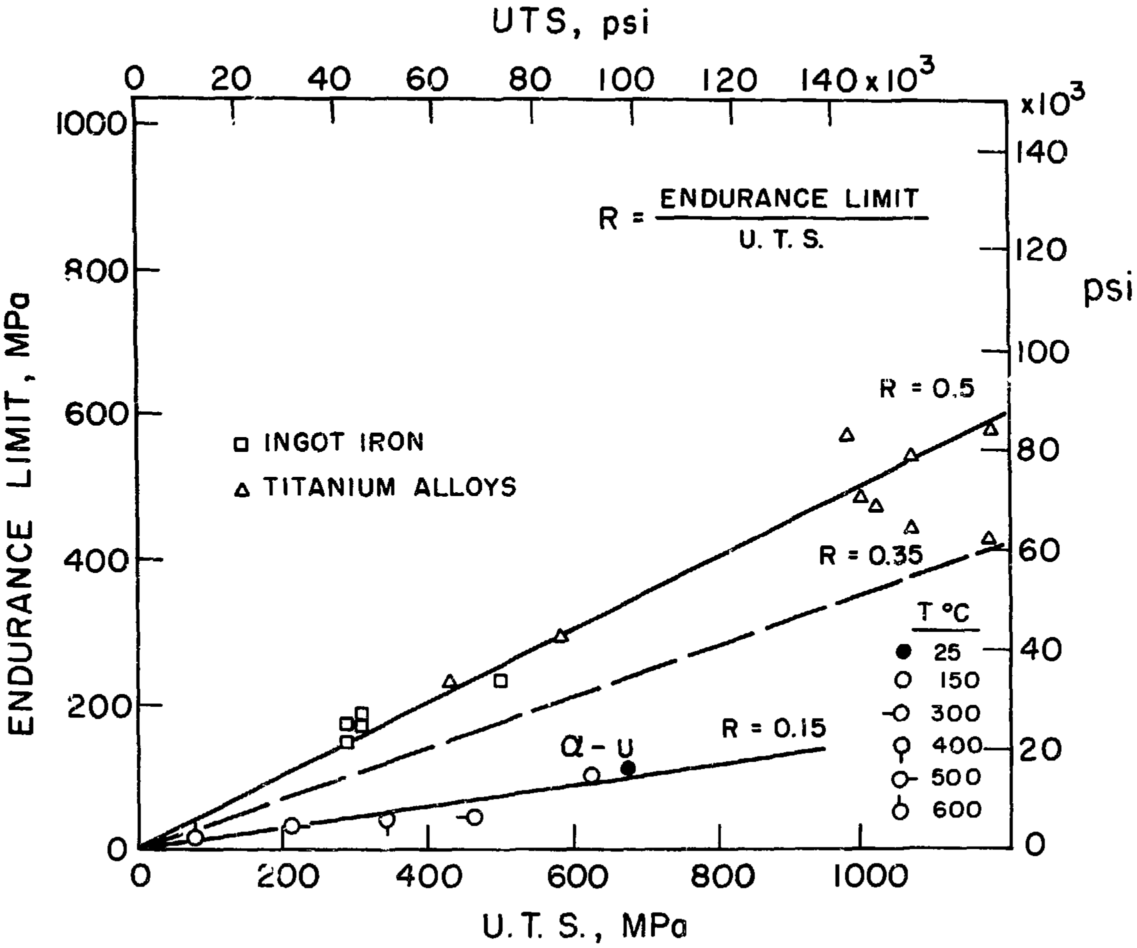

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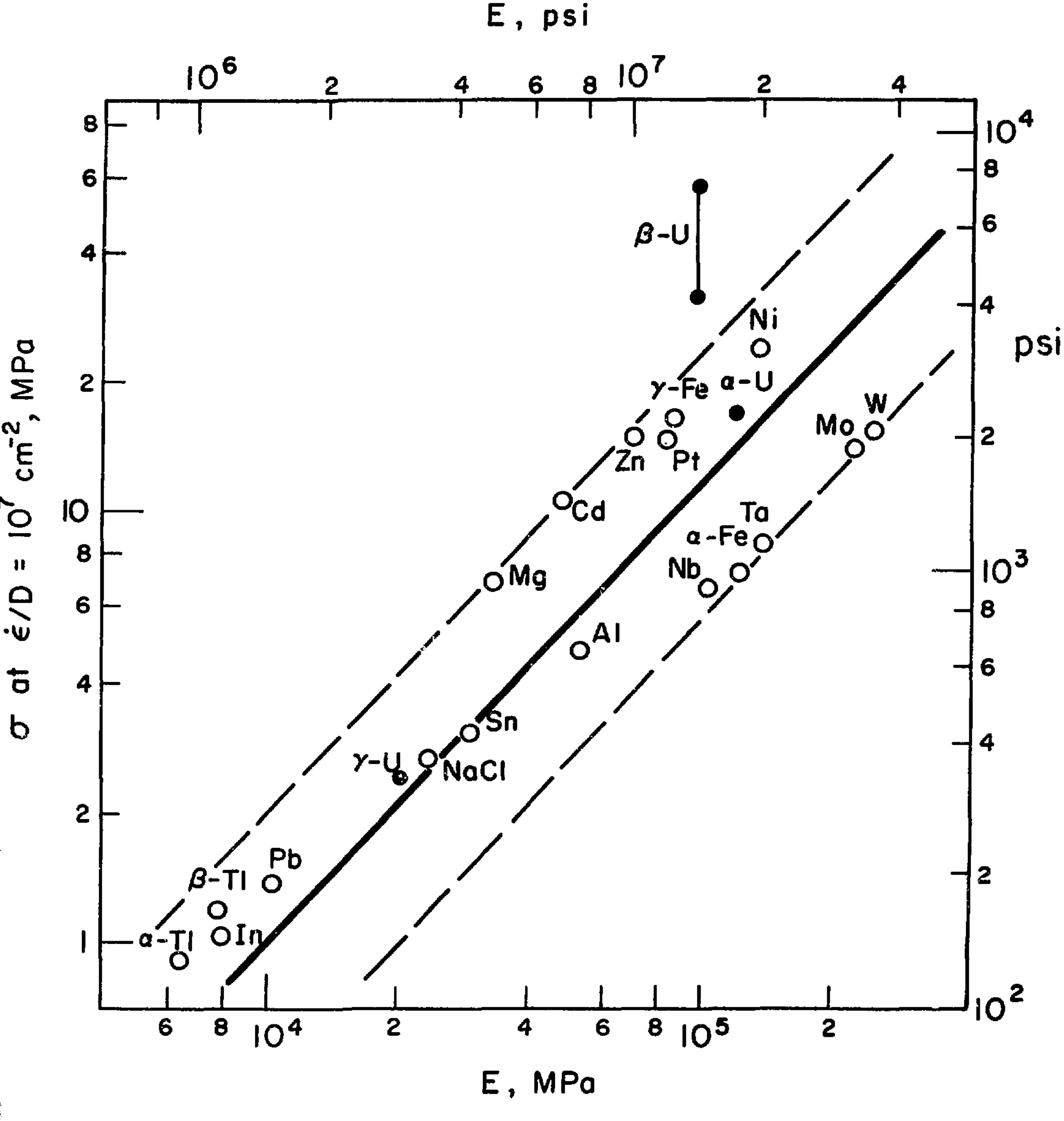

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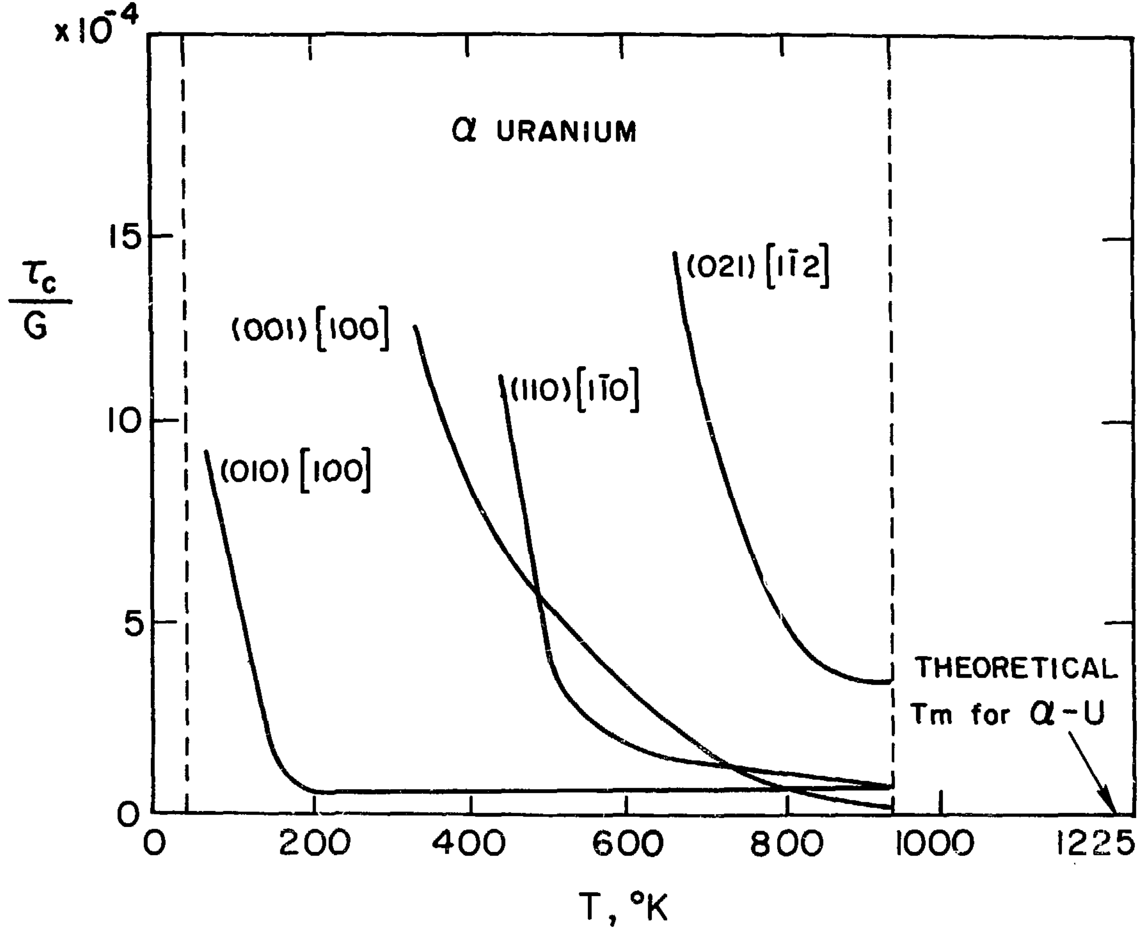

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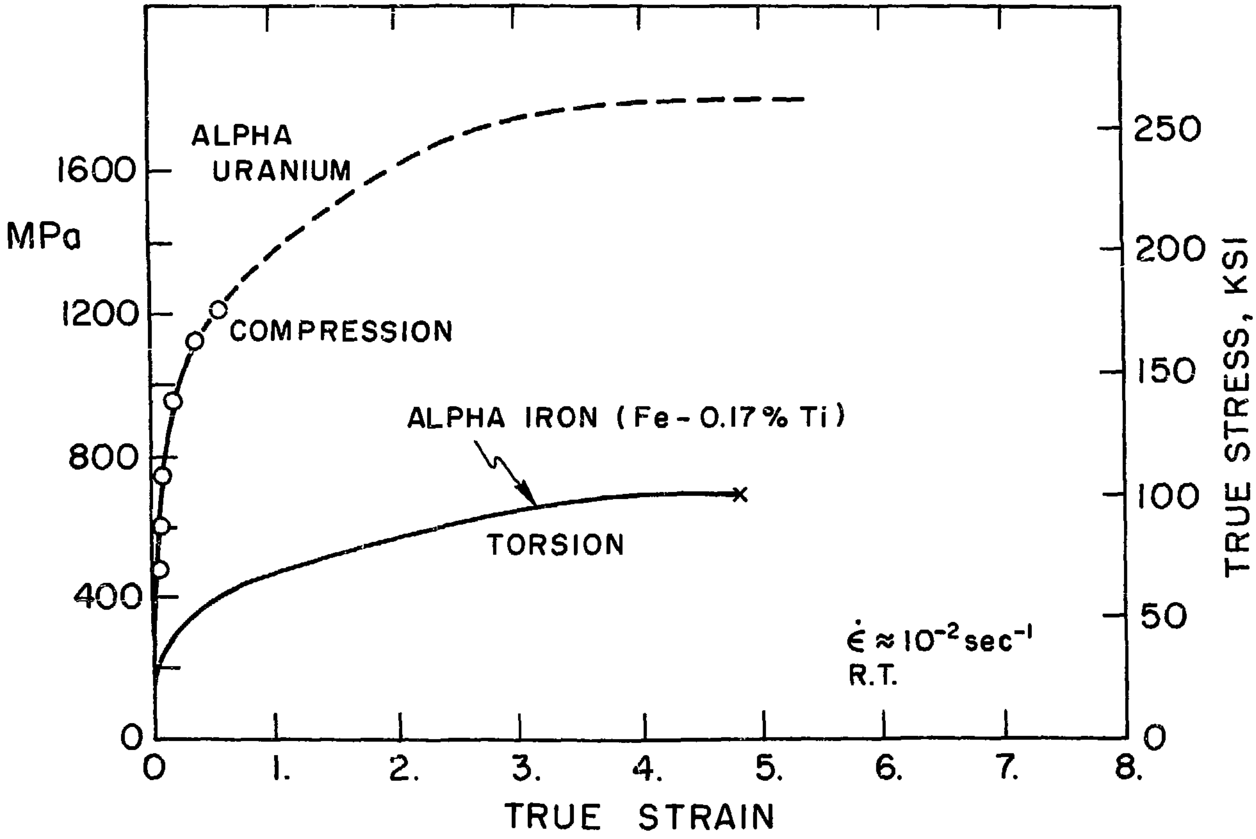

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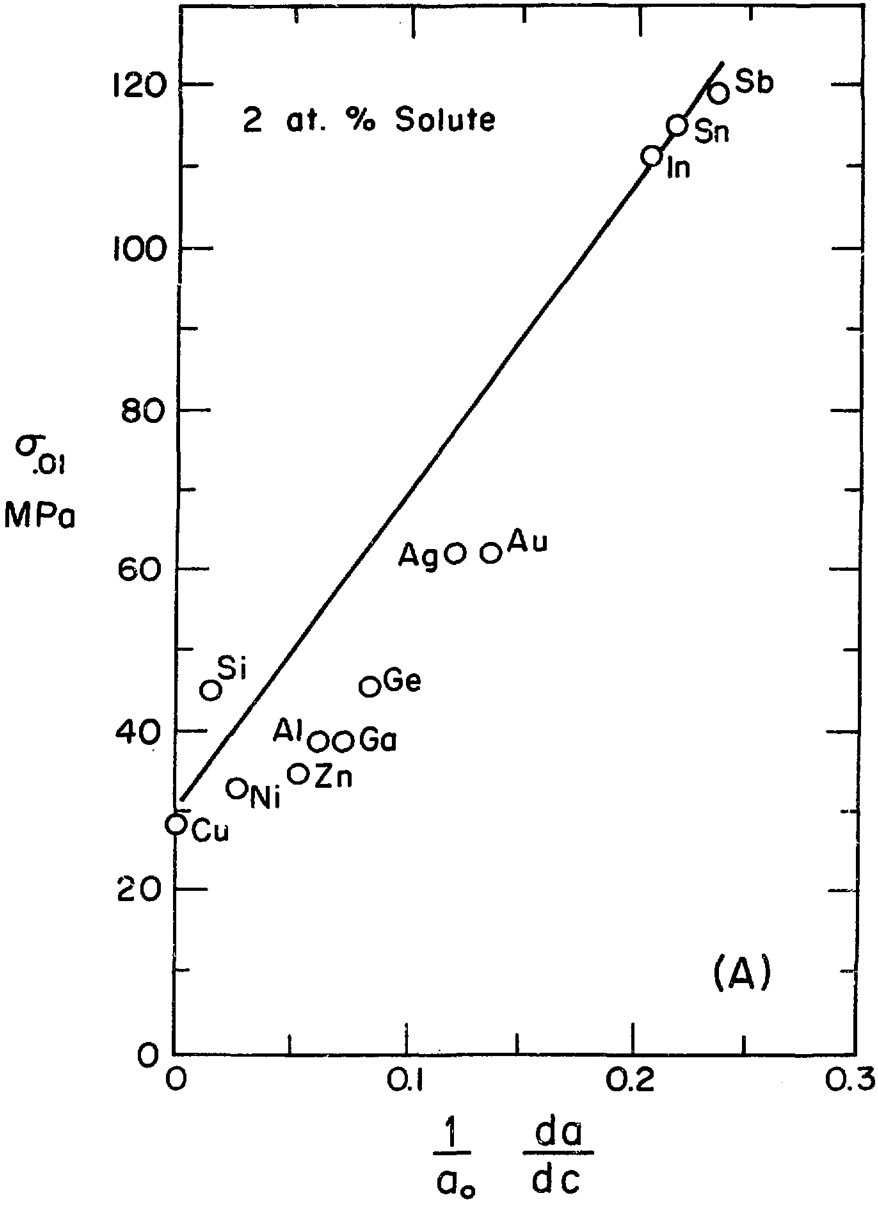

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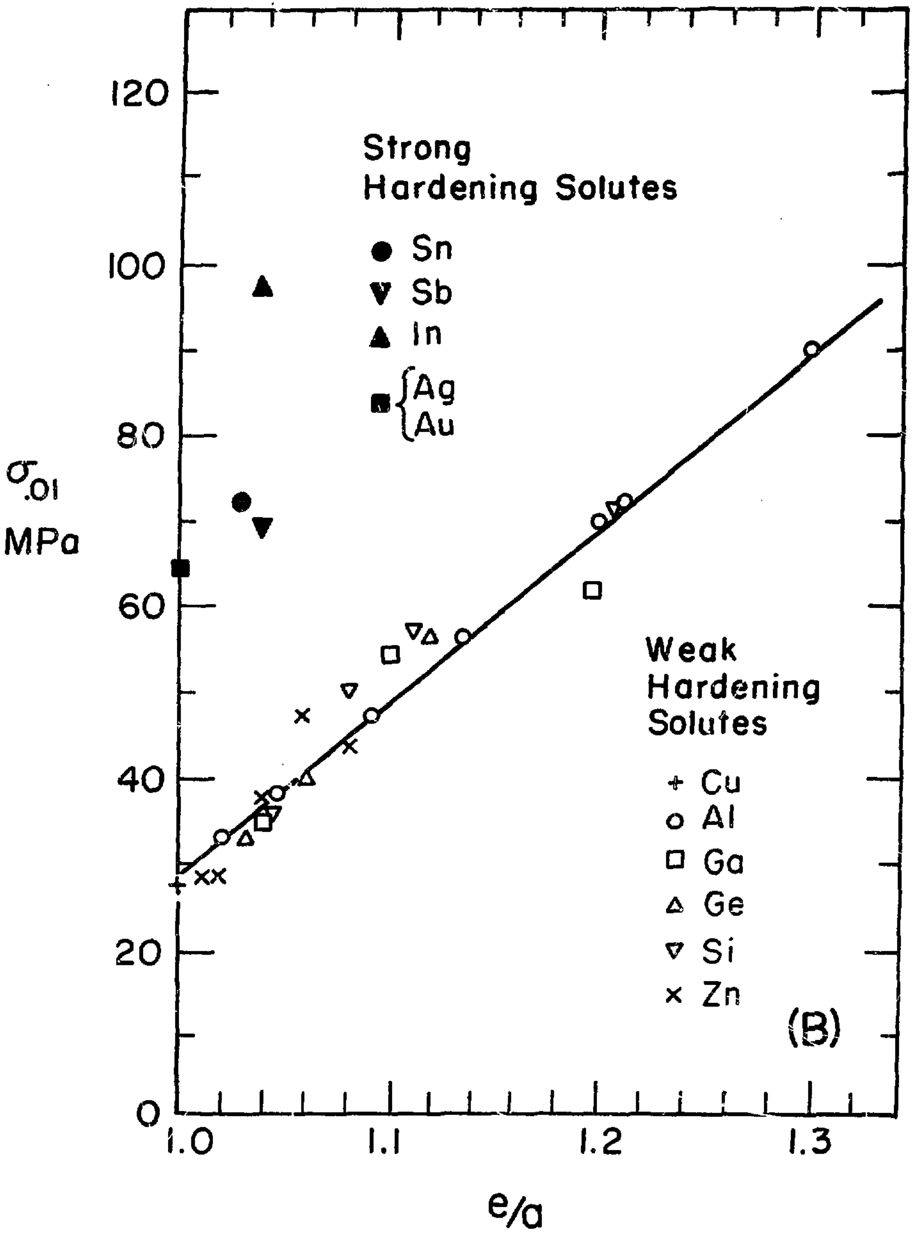
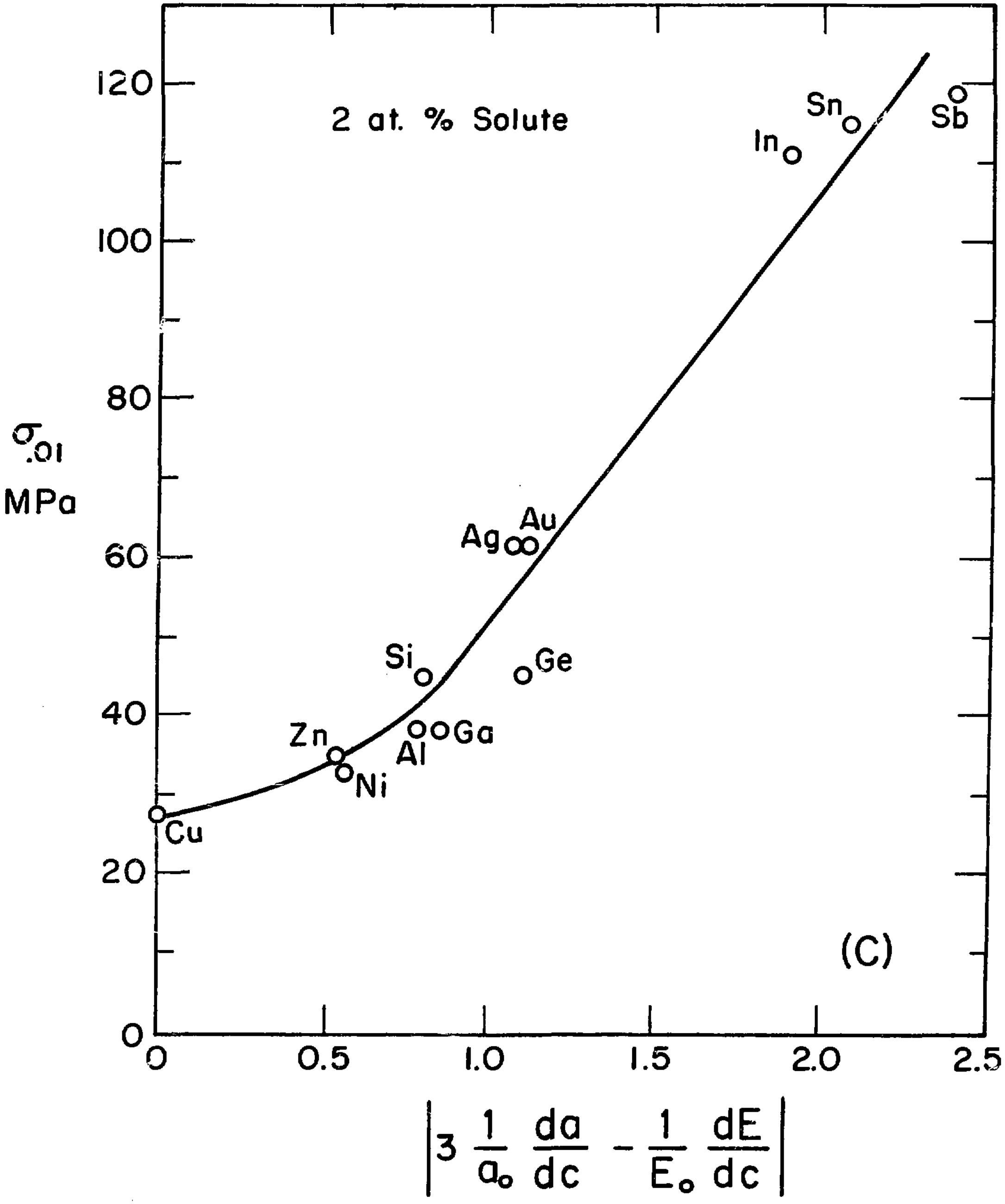
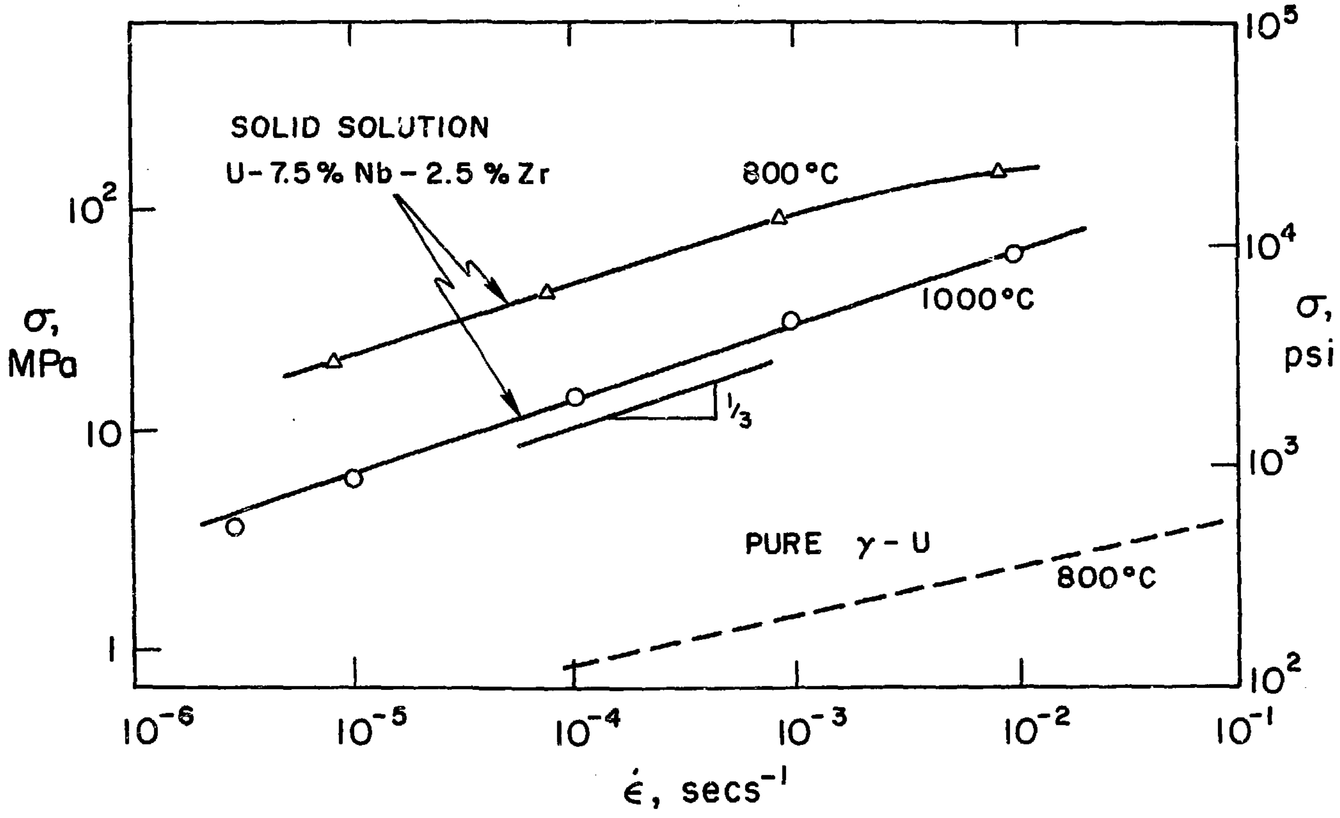

Strong Hardening Solutes
● Sn
▼ Sb
▲ In
■ Ag, Au
Weak Hardening Solutes
+ Cu
○ Al
□ Ga
△ Ge
▽ Si
× Zn
(B)
$\sigma_{.01}$ MPa
0
20
40
60
80
100
120
e/a
1.0
1.1
1.2
1.3

Fig. 9b.



2 at. % Solute
$\sigma_{.01}$
MPa
0
20
40
60
80
100
120
Cu
Zn
Ni
Al
Ga
Si
Ge
Ag
Au
In
Sn
Sb
(C)
0
0.5
1.0
1.5
2.0
2.5
$\left|3\frac{1}{a_o}\frac{da}{dc}-\frac{1}{E_o}\frac{dE}{dc}\right|$

Fig. 9c.



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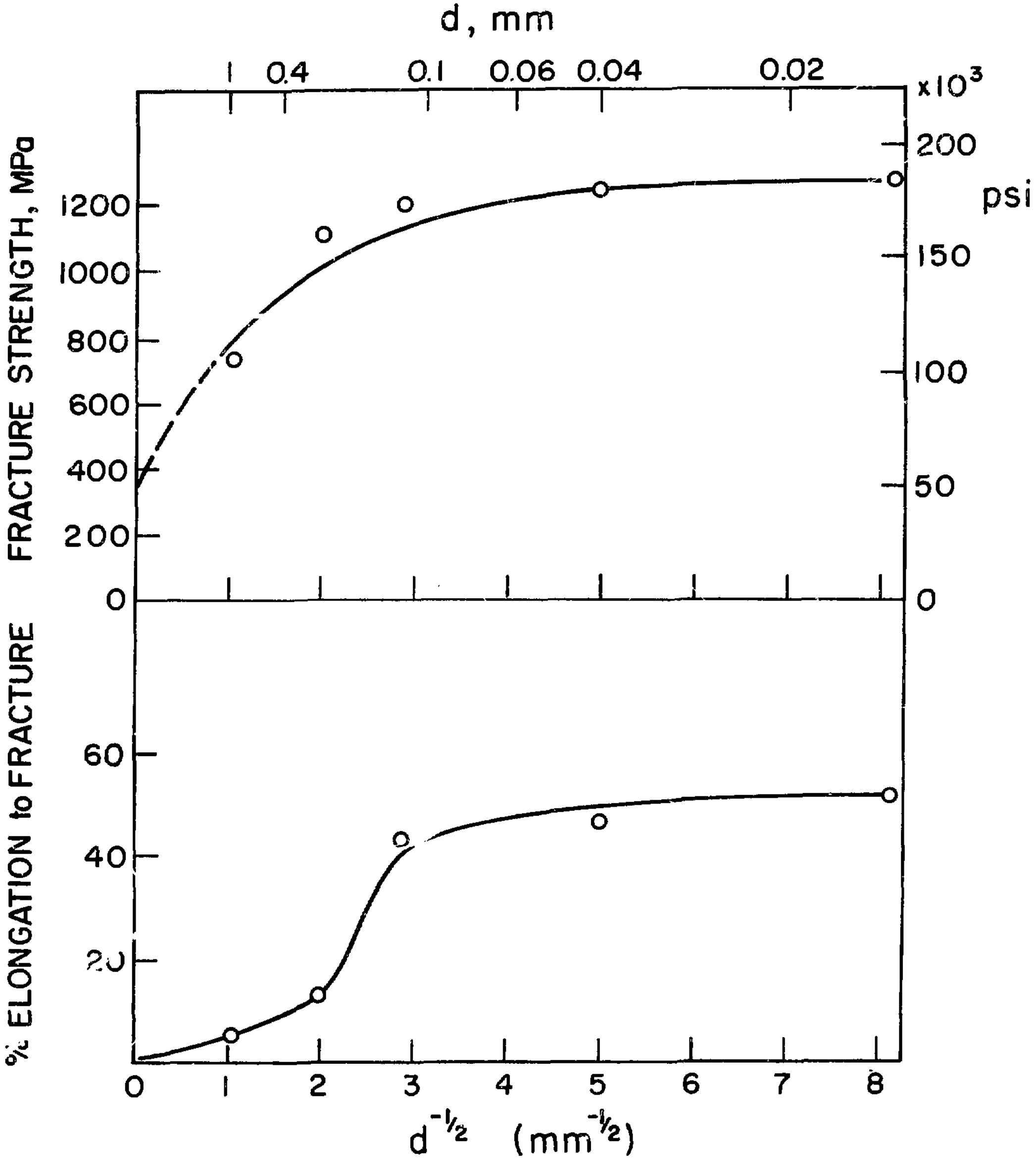

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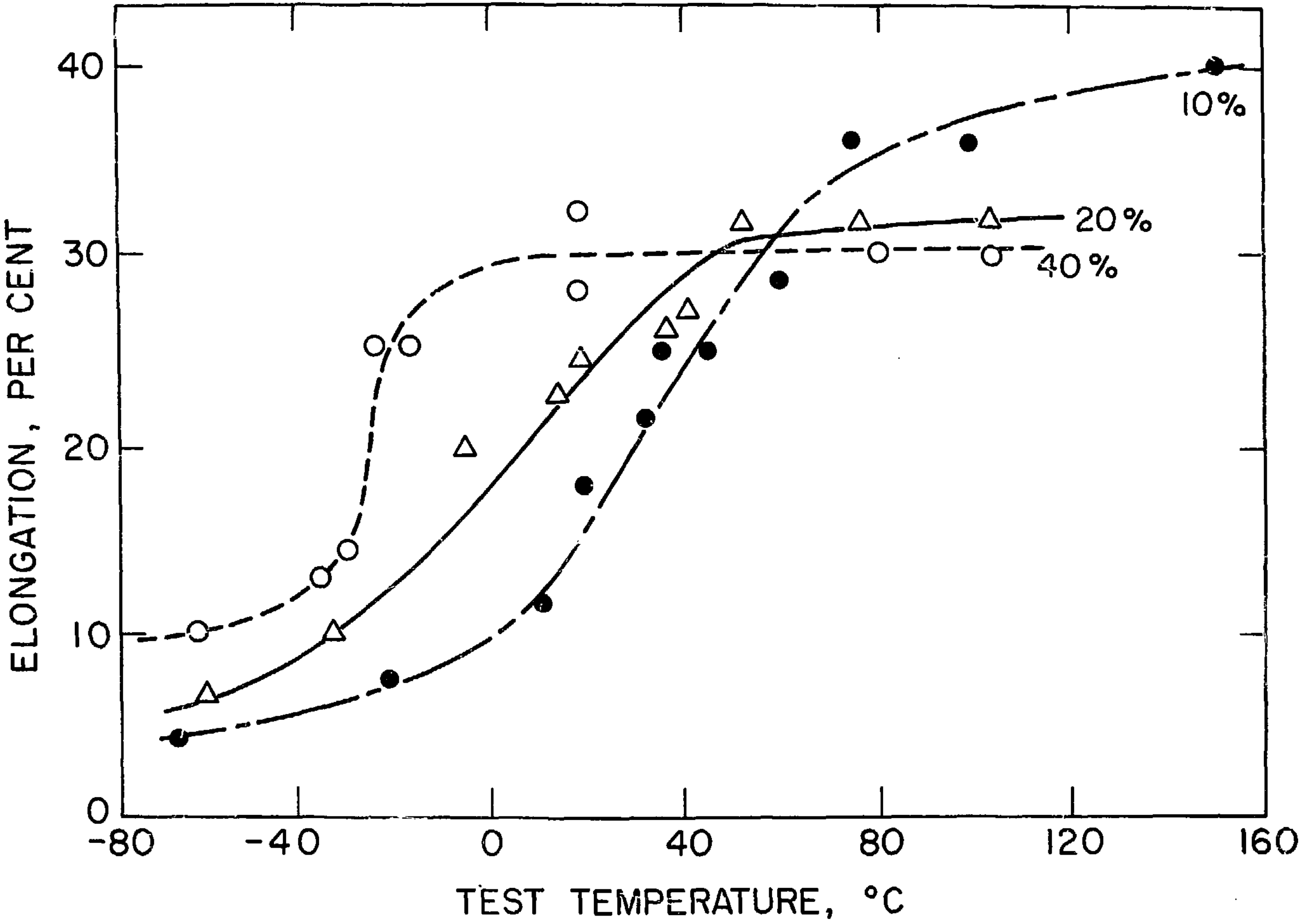

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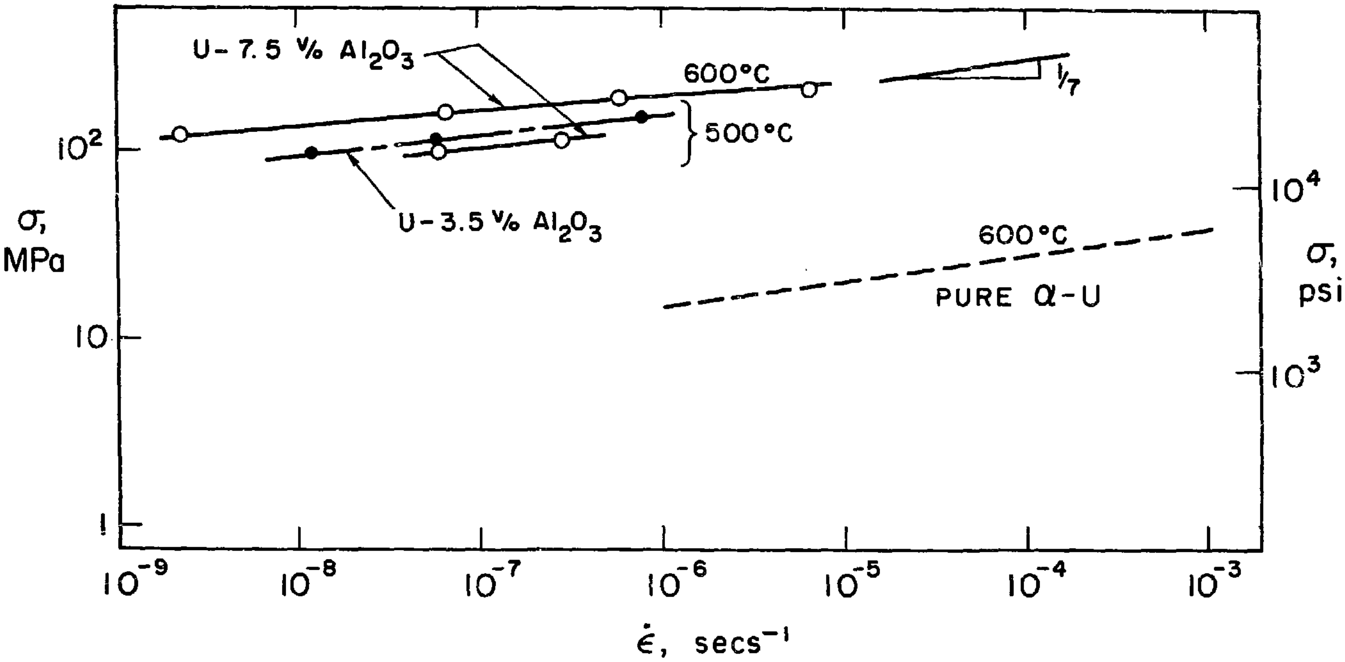

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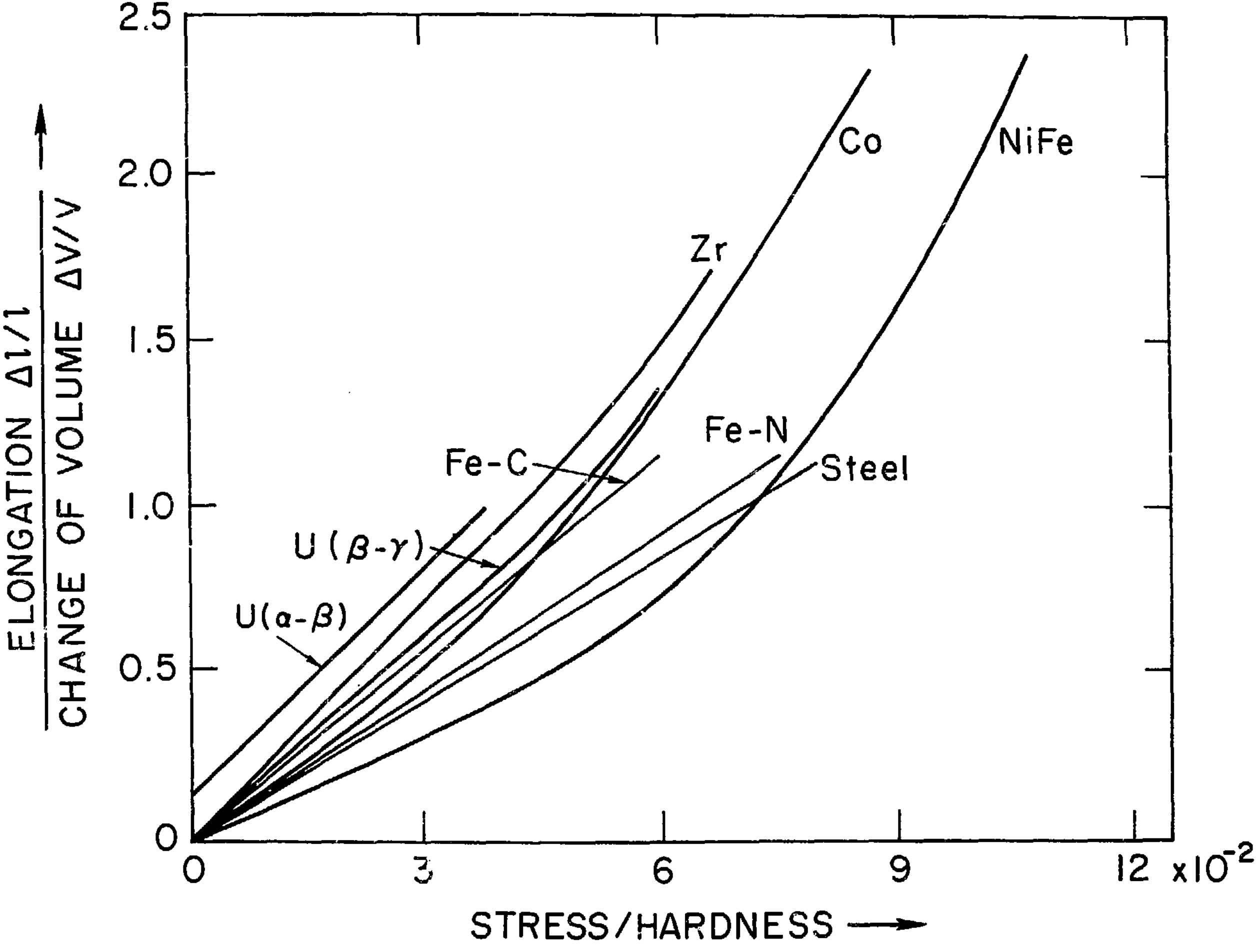

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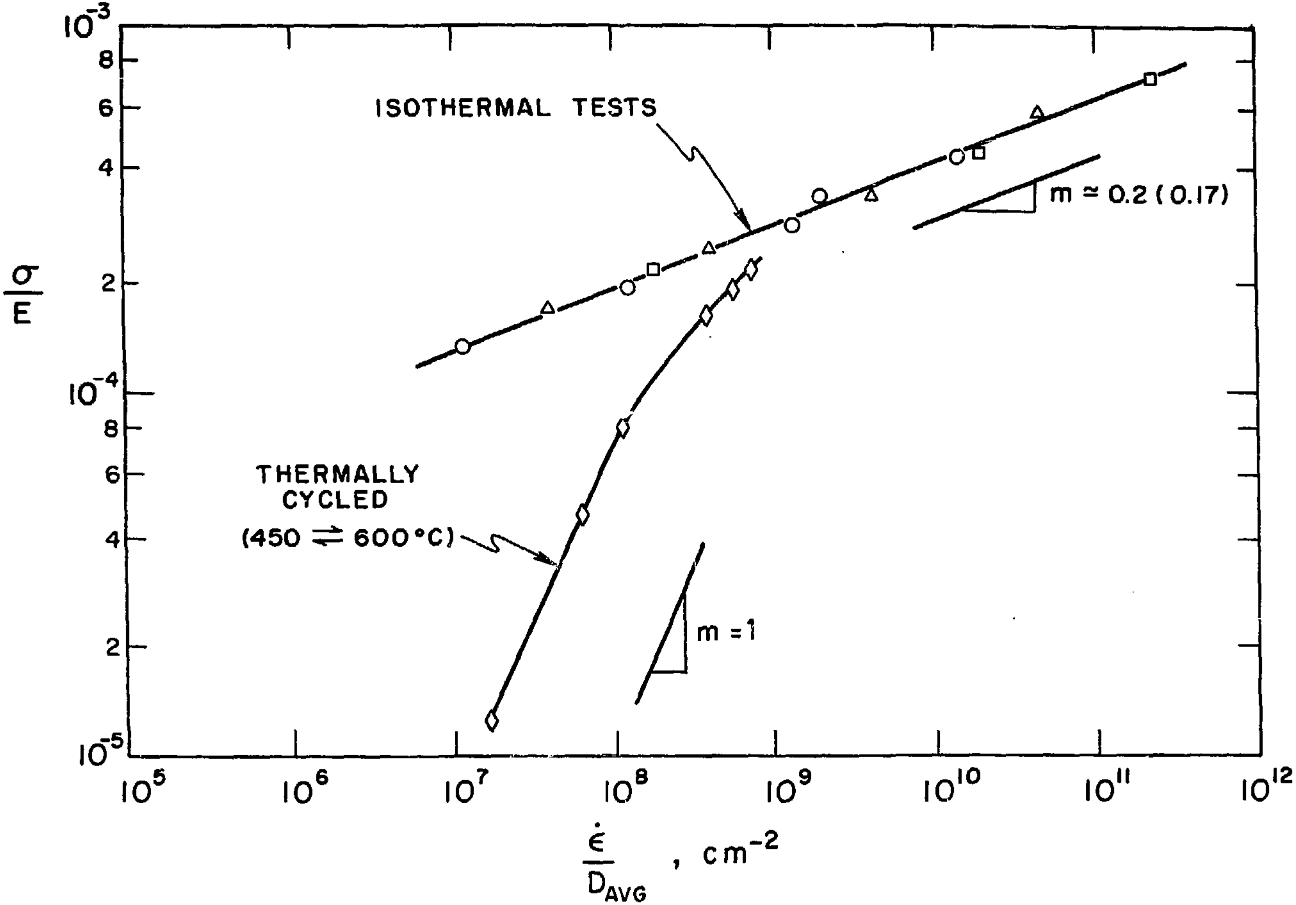

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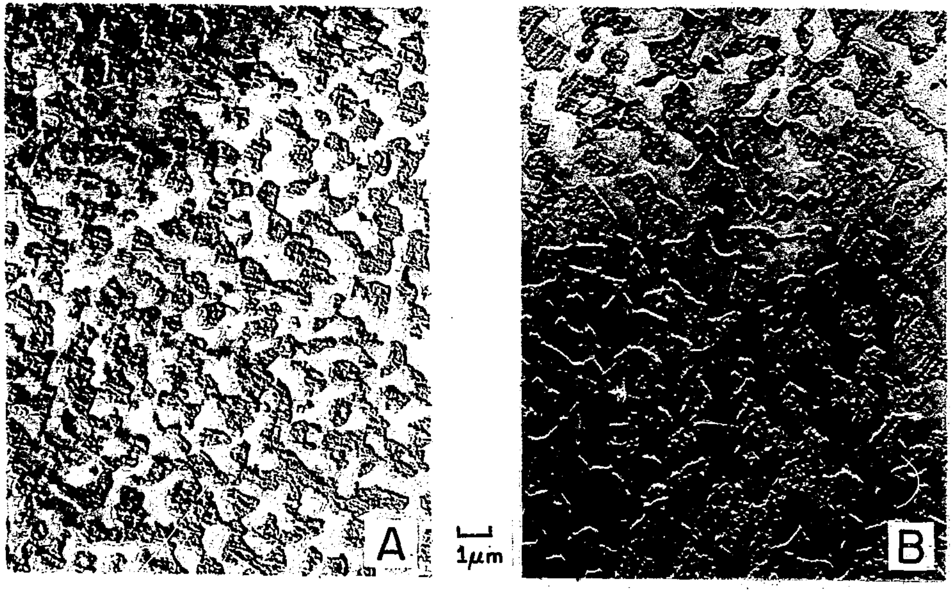

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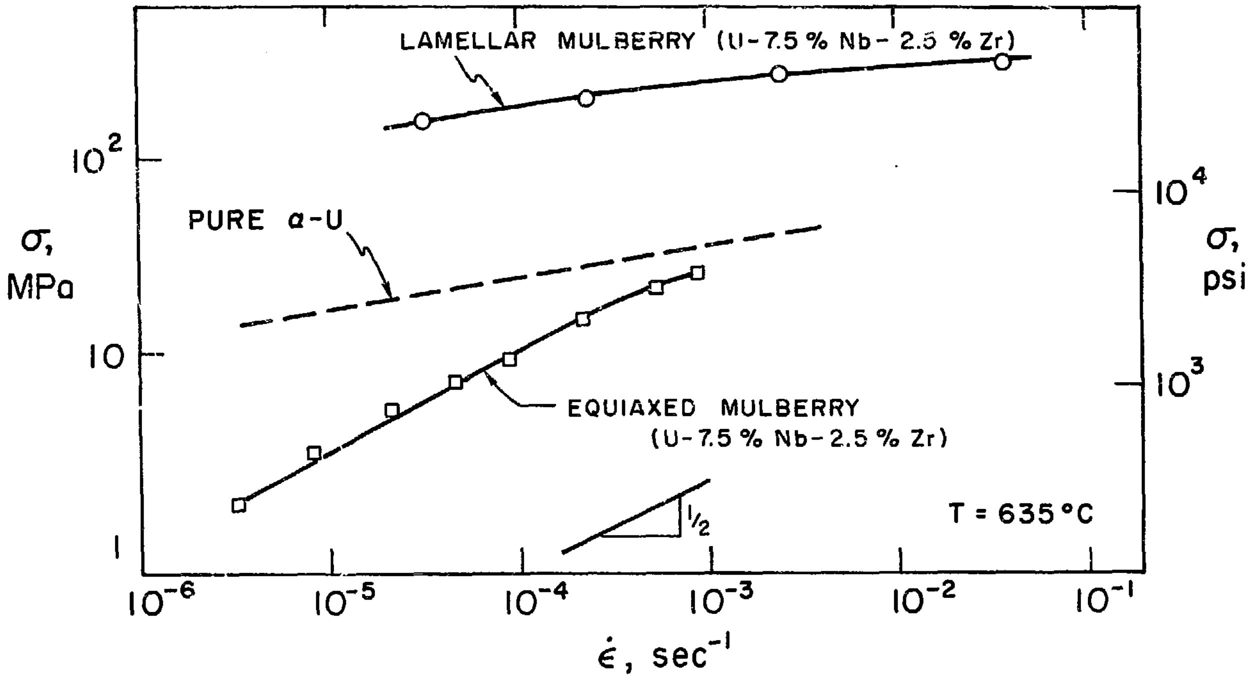

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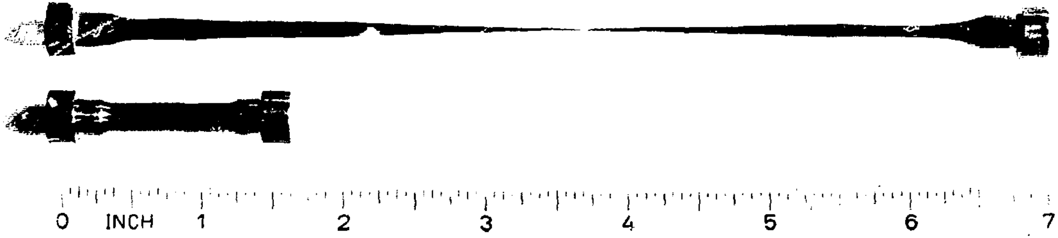

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