THE EFFECT OF HYDROSTATIC PRESSURE ON THE DUCTILE–BRITTLE TRANSITION IN MOLYBDENUM*

J. R. GALLI† and P. GIBBS‡

The ductile–brittle transition temperature for bend specimens of recrystallized powder metallurgy molybdenum has been shown to be lowered from 50°C to 2°C by the application of a hydrostatic pressure of ∼20,000 psi (∼1.4 kbar). The pressure medium was kerosene. The samples were obtained from 0.020 in. dia. wire which was stated to be 99.95% pure and after recrystallization had an average grain diameter of 1/40 mm. It was shown within experimental error that an applied hydrostatic pressure of 20,000 psi results in an increase in the fracture stress by this same amount and thus it is concluded that the rate controlling mechanism of fracture is governed by the tensile component of the applied stress and in fact is independent of a corresponding change in the maximum shear stress of 10,000 psi. It is argued that any critical shear stress which may be important to the fracture process must have a value at or below the value of the maximum shear stress at the transition temperature corresponding to atmospheric pressure. It is estimated that at least for some cases this value is as low as 37,500 psi.

EFFET DE LA PRESSION HYDROSTATIQUE SUR LA TRANSITION DE RUPTURE (DUCTILE–FRAGILE) DU MOLYBDENE

Les auteurs montrent que la température de transition entre la rupture ductile et fragile par flexion d'éprouvettes de molybdène fritté et recristallisé, passe de 50 à 2°C, lors de l'application d'une pression hydrostatique de 20.000 psi (14 kg/mm²). Dans cette étude, la pression était exercée par l'intermédiaire de kérosene. Les échantillons provenaient de fils 0,51 mm. de diamètre à 99,95% de pureté, présentant après recristallisation une grosseur de grain de 1:40 mm.

Les auteurs montrent également qu'à l'erreur expérimentale près, l'application d'une pression hydrostatique de 20.000 psi (14 kg/mm²) entraîne un accroissement du même ordre de grandeur de la charge de rupture. Ils en concluent que la composante de traction de las pression appliquée gouverne le mécanisme de contrôle de la vitesse de rupture et que ce mécanisme est le fait indépendant d'une variation correspondante de la cission critique maximale de 10.000 psi (7 kg/mm²). Les auteurs démontrent que toute tension critique de cisaillement susceptible de jouer un rôle important dans le processus de fracture doit avoir une valeur égale ou inférieure à la cission maximale pour la température de transition relative à la pression atmosphérique. Ils estiment que la valeur de celle-ci est, du moins dans certains cas, aussi faible que 37.500 psi (20 kg/mm²).

DER EINFLUß HYDROSTATISCHEN DRUCKS AUF DEN ÜBERGANG DUKTIL–SPRÖDE VON MOLYBDÄN

Bei Anwendung eines hydrostatischen Druckes von ∼20,000 psi (∼1.4 kbar) ergab sich eine Erniedrigung der Übergangstemperatur des Übergangs duktil–spröde bei Beigung von Proben aus rekristallisiertem gesintertem Molybdän von 50°C auf 2°C. Als Druckmedium wurde Kerosin benutzt. Die Proben wurden aus Draht von 0.020 in. Durchmesser mit einem angegebenen Reinheitsgrad von 99.95% hergestellt; nach der Rekristallisation hatten sie einen mittleren Korndurchmesser von 1/40 mm. Innerhalb der experimentellen Fehler ergab ein hydrostatischer Druck von 20,000 psi eine Erhöhung der Bruchspannung um denselben Betrag; wir schließen daher, daß der geschwindigkeitsbestimmende Bruchmechanismus durch die Zugkomponente der äußeren Spannung bestimmt ist und tatsächlich unabhängig von einer entsprechenden Änderung der maximalen Scherspannung um 10,000 psi ist. Es wird begründet, daß jede kritische Schubspannung, die für den Bruchprozeß wesentlich sein soll, einen Wert haben muß, der dem Wert der maximalen Schubspannung bei der Übergangstemperatur, die bei Atmosphärendruck auftritt, gleichkommt oder unter ihm liegt. Es wird geschätzt, daß zumindest in einigen Fällen dieser Wert nicht höher als 37,500 psi ist.

INTRODUCTION

It is generally accepted that the ductile–brittle transition is due to the competition between yield and fracture, and that a strongly temperature dependent yield stress is a prerequisite for a narrow transition region in b.c.c. metals. Most of the widely recognized theories of the ductile–brittle transition have in common the need for the operation of slip dislocations which can subsequently result in either fracture or yield, depending upon the temperature and other important variables.[1–7] Cottrell[1,2] and Petch[3,5] have independently concluded that the fracture process consists essentially of two parts: (a) the formation of a crack nucleus as a result of dislocation coalescence, and (b) the propagation of this crack. They further conclude that the growth of the crack is more difficult than its initiation. In support of this, Cottrell cites evidence that the hydrostatic component of the applied stress system plays a role in determining the relative brittleness of a material.

EXPERIMENTAL PROCEDURE

The pressure vessel was similar to that described in Refs. 8 and 9. The relative viscosity indicated that the kerosene sample environment was liquid within the range of temperatures and pressures explored. The pressure was determined by measuring the pressure in the hydraulic lines on a precision Heise pressure gauge and then converting this reading to the pressure in the vessel by using the appropriate areal conversions. Frictional forces were calibrated, and found to be negligible below 20,000 psi (∼1.4 kbar) and above this pressure they were less than 1,000 psi (∼0.07 kbar) for all pressures studied.[10]

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† The research is a part of a dissertation submitted for Ph.D. in Physics at University of Utah, Salt Lake City, Utah.
Now at: Department of Physics, Weber State College, Ogden, Utah.
‡ Department of Physics, University of Utah, Salt Lake City, Utah.

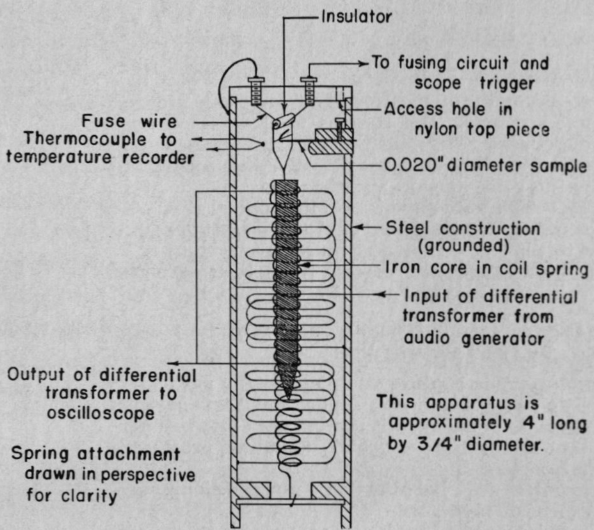

FIG. 1. Schematic of the stressing apparatus.

The temperature was controlled by immersing the pressure vessel in a water bath. The temperatures inside the pressure vessel were measured by a chromel–alumel thermocouple and Bristol Dynamaster Recorder, and compared with those of a thermocouple and a thermometer which were immersed in the temperature bath. The readings differed by less than 1/2°C.

The stressing apparatus (shown schematically in Fig. 1) fit inside of the pressure vessel. When the desired conditions of pressure and temperature were reached, the fuse wire was melted. The stretched spring then applied sufficient stress to the sample to cause failure and the spring (with iron core attached) moved down through the coils of a differential transformer.[10] The output of the transformer was fed into an oscilloscope which was triggered by the melting fuse wire. Thus, deflection was measured as a function of time by recording the scope sweep on film.

The samples were forced to bend around a mandrel of radius $R = 0.046$ in., and the maximum plastic strain was observed to be constant for all samples tested. The strain ϵ in the outer fiber of a cylindrical sample of radius $r = 0.01$ in. is thus $\epsilon = r/(r + R) \simeq 18\%$.

At fracture, the electrical circuit through the sample, chassis, spring, and connecting parts was broken, and a voltage "kick" was observed in the output of the transformer.

The samples were cut into 3/4 in. lengths from 20 mil (99.95 + %) powder metallurgy molybdenum wire (purchased from the Fansteel Corporation). A sharp 1/16 in. bend was made at one end to serve as an attachment for the stressing spring. A 1-hr vacuum anneal at 1500°C resulted in an average recrystallized grain size of ~1/40 mm. The reported principal impurities (in per cent) were: C—0.005, O—0.004, N—0.001, W—0.008, Si—0.005, Ni—0.003, Cr—0.001, Ca—0.001, Cu—0.001, Ti—0.01, Sn—0.01, Mg—0.001.

The deflection rate was essentially independent of temperature and pressure (40 in./sec for the first 4 msec). After 4 to 5 msec, the deflection rate approached zero smoothly, after which the system remained essentially at rest. The maximum angle through which the ductile samples were observed to bend was ~60°.

The deflection of the sample due to contact with the mandrel would be expected to follow the geometrical relationship[10]

$$\frac{d\theta}{dt} = \frac{dx/dt}{[L - (R + r)\theta] \cos \theta}$$

where θ is the angle through which the rod is in contact with the mandrel, x is the deflection of the spring, and L is the original length of the lever arm (1/4 in.). From the above equation the "angular velocity" was calculated to increase from 160 rad/sec to 417 rad/sec in going from 0° to 60°. Assuming the strain rate is proportional to the angular velocity,[10] the strain rate increased by this same factor (2.6) during any given run. Although the exact value of the strain rate could not be determined exactly, it was assumed to be the same for all tests at the same sample deflection. The *maximum* strain rate was roughly estimated[10] to lie between 40 sec^{-1} and 40×10^3 sec^{-1}.

EXPERIMENTAL RESULTS

Deflection commenced immediately upon application of stress. The fracture usually occurred later by a time interval which depended upon both temperature and hydrostatic pressure (with a scatter of about 1–3 msec). At 25°C the average time to fracture increased from 2 msec at atmospheric pressure to 4 msec at 15,000 psi (~0.95 kbar). At 2°C the average time varied from 0.5 msec at atmospheric pressure to 4 msec at 27,500 psi (~1.9 kbar). At atmospheric pressure the average time varied from 0.5 msec at 2°C to 4 msec at 55°C. This might arise because the local strain rate increases with deformation in this geometry.

Considering the samples which hadn't broken after 4 msec to be ductile produces the transition region shown in Fig. 2. The black portion of the circles represented the fractions of six samples fractured according to this criterion. The 4 msec interval was

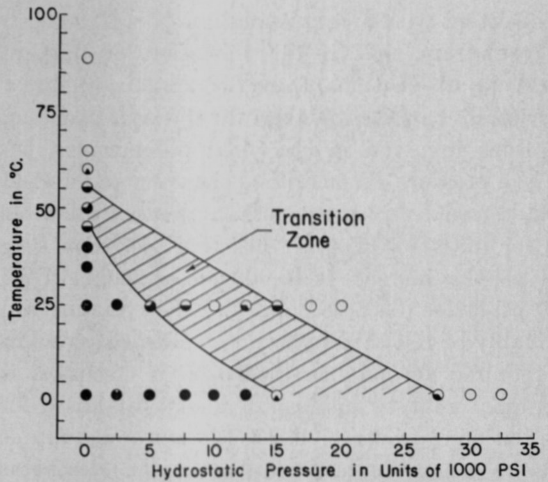

FIG. 2. Temperature and pressure effect on fracture in the transition region. The black portions of the circles represent the fractions of six samples which fractured before 4 msec.

also significant since afterwards the deflection rate started to decrease.

If the transition is defined to occur at the mid point of the transition region, then the transition temperature at atmospheric pressure is ~50°C, that at ~10,000 psi is 25°C, and that at ~21,000 psi is 2°C. However, it should be remembered that pressures above 20,000 psi may be as much as 1,000 psi below the reported value because of the friction between the piston and cylinder as discussed earlier.

DISCUSSION

The results of the present experiment will be compared to those of the more common tensile experiments, since the ratio of the maximum tensile stress σ_m to maximum shear stress τ_m is equal to 2 for both the bend and the tensile tests, and apparently the transition temperatures are essentially the same.

The slope of yield strength vs. temperature curves in the present studies is probably similar to that of Bechtold,[11,12] Fig. 3. The fracture stress is almost independent of temperature for the ranges of interest.[12] The slope of the yield stress curve over this temperature range has essentially the same slope for ASTM grain size No. 5.9 (~500 grains/mm^2) as ASTM grain size No. 7.8 (~1800 grains/mm^2).[12] Bechtold's data refer to 900 grains/mm^2 while the authors' refer to 1600 grains/mm^2. The effect of impurities on the 0.2% yield strength of tantalum (which has similar yield characteristics as molybdenum) is rather large but the slope appears to be changed only slightly over relatively wide ranges of impurity concentrations.[13] As can be seen from Bechtold's data for molybdenum the slope at both 0°C and 50°C is essentially the same over 3 orders of magnitude change in the strain rate. Bridgman[14] found pressure to have negligible effect on the yield stress of molybdenum below 120,000 psi (~8.5 kbar).

All evidence indicates that the required applied stress to cause fracture is raised by the application of a hydrostatic pressure and in fact the change in the fracture stress which is required to lower the transition temperature by 50°C is just equal to the applied hydrostatic pressure of ~20,000 psi as reference to Fig. 3 will indicate. (It is assumed that the yield curves can be extrapolated 50°C below the intersection between yield strength and fracture strength.) It should, however, be pointed out that the transition criterion previously discussed for this experiment involves approximately an 18% strain in the outer fiber so that this transition temperature lies slightly to the right of the intersection between the fracture stress and yield stress curves. However, if this shift is not a function of pressure it will not be important to the discussion. Studies using a smaller strain criterion could further clarify this point.

The effect of applying a hydrostatic pressure P to a homogeneous cubic crystal is to add to any tensile stresses, which might originally be present, a stress of magnitude $-P$ and to leave the shear components unaltered. This result was used in this investigation since the sample material consisted of relatively pure (99.95%) powder metallurgy recrystallized molybdenum of b.c.c. structure and was believed to contain no gross inhomogeneities.

Since the number used to indicate the fracture stress (or yield stress) is the maximum tensile component of the applied stress system (not including hydrostatic pressure) and since the effect of hydrostatic pressure is to reduce all tensile components including the maximum value by the amount of the

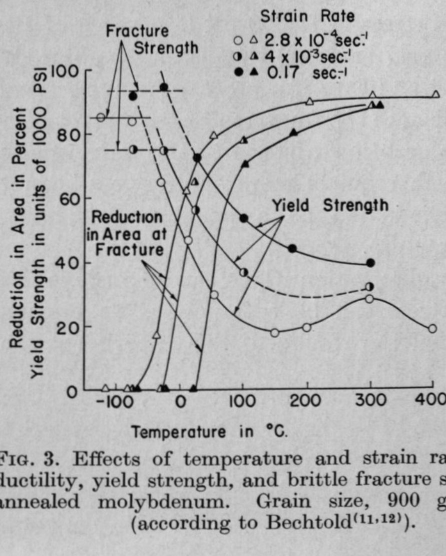

FIG. 3. Effects of temperature and strain rate on the ductility, yield strength, and brittle fracture strength of annealed molybdenum. Grain size, 900 grains/mm^2 (according to Bechtold[11,12]).

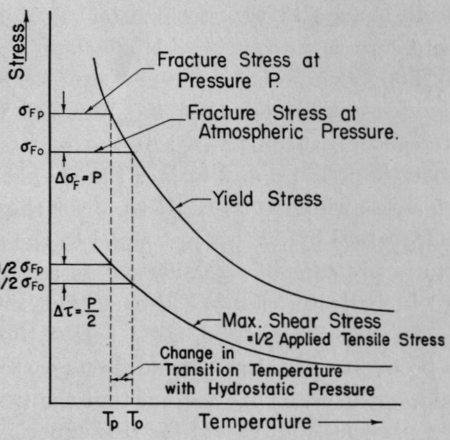

FIG. 4. Schematic representation of the fracture stress, yield stress, maximum shear stress, and the effect of applying a hydrostatic pressure.

applied hydrostatic pressure, and since it is then necessary to increase this maximum tensile stress by precisely this same amount before fracture will occur, it appears that fracture cannot occur until the maximum tensile stress achieves a critical value independent of the increase in shear stress which also occurs when the applied stress is increased. These ideas are illustrated graphically in Fig. 4.

The required fracture stress is independent of shear stress over the range studied. Thus if shear stresses play a role in the fracture process (for example by nucleating a crack which is subsequently controlled by tensile stresses) it must be that this occurs at or below the maximum shear stress present at atmospheric pressure (1/2 the atmospheric fracture stress at the transition temperature). The fracture stress measured for a variety of experiments dealing with the ductile–brittle transition of molybdenum appears to range between 75,000 psi and 120,000 psi with most of the values below 100,000 psi.[12,15,16] When the fracture stress is 75,000 psi (the value of Bechtold's "weakest" samples) the maximum shear stress is 37,500 psi and thus if a process involving shear is going to influence the fracture process it must be able to occur at or below a stress of this value and any process which requires more shear stress than this must be eliminated as a possible candidate for initiating fracture.

A further observation which can be made is that if the yield stress can be extrapolated as previously assumed (and the results of this experiment are *at least* consistent with an extrapolation of ~20,000 psi) then it appears that if the initiation of fracture involves slip dislocations, these dislocations require a smaller stress to function than do the corresponding dislocations involved in the yield phenomenon because at a pressure P the critical shear stress for yield is an additional $P/2$ greater than the critical shear stress for initiating fracture (see Fig. 4). For $P =$ 20,000 psi this increase is 10,000 psi or 26.6% of the 37,500 psi stress discussed above. Since fracture can conceivably be initiated by twinning at a stress below that required for slip as Sleeswyk[17] discusses, it would seem that twinning is a likely candidate for the initiation of fracture. These results would appear to require generation and propagation of microtwins to take place below about 37,500 psi if they are responsible for fracture nucleation. However, the growth of the fracture appears to be the rate limiting process since fracture cannot occur until a critical tensile stress is reached.

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