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2002 J. Phys.: Condens. Matter 14 10907

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# Pressure strengthening and its application to the analysis of hydrogen sample-radius behaviour in a tungsten gasket as a function of the initial sample-radius-to-tip-radius ratio

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Received 10 June 2002

Published 25 October 2002

Online at [stacks.iop.org/JPhysCM/14/10907](http://stacks.iop.org/JPhysCM/14/10907)

## Abstract

By calculating the single-crystal elastic constants of W and Mo by first-principles methods the pressure strengthening of the yield stress was obtained. This was used to calculate the behaviour of the hydrogen sample hole size in a tungsten gasket. It was found that drastic changes occur depending on  $r_s^0/r_t$  where  $r_s^0$  is the initial sample radius and  $r_t$  is the diamond tip radius. It is seen why, with  $r_s^0/r_t = 0.9$ , the Carnegie group cannot and have not exceeded pressures of 230 GPa and why, with  $r_s^0/r_t = 0.5$ , the Cornell group has been able to reach 342 GPa.

## 1. Pressure strengthening

Pressure strengthening makes possible the attainment of multimegabar pressures in the diamond anvil cell [1–5]. Its existence was first noted in a paper given by the present author in 1972 [6].

The first accurate measurement on the effect of pressure on yield strength was made by Chua and Ruoff in 1973 [7]. This along with the theories of yield strength [7] enables one to conclude that the yield strength of an equiaxed polycrystalline aggregate scales with pressure as follows:

$$\sigma = \frac{G/(1-\nu)}{G_0/(1-\nu_0)}\sigma_{00} = F(P)\sigma_{00} \quad (1)$$

where  $\sigma_{00}$  is the yield strength at atmospheric pressure,  $G$  is the shear modulus under pressure,  $\nu$  is Poisson's ratio under pressure,  $G_0$  and  $\nu_0$  are the corresponding values at atmospheric pressure and  $F(P)$  is the pressure strengthening factor.

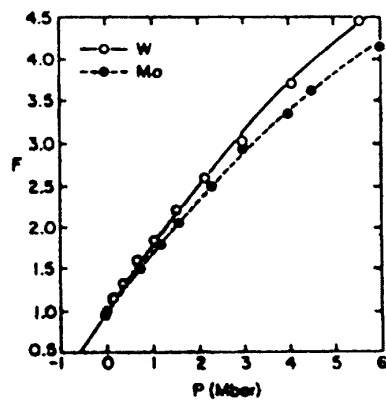


Figure 1. The pressure strengthening factor,  $F$ , versus pressure for W and Mo.

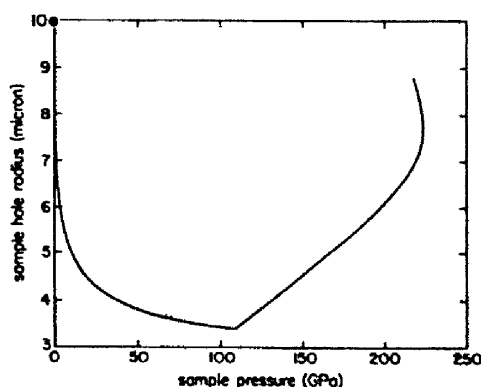
One way of obtaining an approximation for  $F(P)$  is to use the ultrasonically measured single-crystal elastic coefficients at atmospheric pressure and at moderate pressures. They can be connected to  $G$  and  $\nu$ , using well known methods [8, 9], so that an expression linear in  $P$  is obtained for  $F(P)$ . In the case of diamond,  $F(P) \approx 2$  at  $P \approx 5$  Mbar [10].

Theoretical methods can also be used to obtain for cubic crystals such as Mo and W the individual elastic constants  $B$ ,  $(C_{11} - C_{12})/2$  and  $C_{44}$  and hence the exact variation with pressure of  $F(P)$ . With this development of modern computational techniques, it is certainly true that  $F(P)$  can be calculated with substantially greater accuracy in the megabar regime [11] than it can be measured experimentally at this time [13]. Such measurements are very difficult. The individual elastic constants were computed using the local density approximation of density-functional theory with a full-potential implementation of the linear muffin-tin orbital method to calculate the total energies of the crystal in various strain states. For W, a scalar relativistic version to the LMTO scheme was used. This included relativistic shifts, but spin-orbit splitting was omitted. The elastic constants obtained were then used to compute  $G$  (and since  $B$  is known,  $\nu$ ) using the method of Hashin and Strikmann [8]. The results are shown in figure 1. Note that the pressure strengthening factor is approximately 4.5 at six megabars [11, 12]. It is this which makes possible the attainment of a pressure of 416 GPa [4], greater than that at the Earth's core (361 GPa) and 560 GPa [5] (approaching the TPa regime).

## 2. Analysis of sample-radius variation of hydrogen in a tungsten gasket

It is of interest to combine the effects of strain hardening and pressure strengthening to analyse the situation for a sample of hydrogen in a gasket at high pressure. The sample is located in a circular hole whose centre is at the tip centre. The problem is to calculate the sample radius as a function of pressure for different initial ratios of sample radius ( $r_s^0$ ) to tip radius ( $r_t$ ), i.e.  $r_s^0/r_t$ . The diamond was described using nonlinear finite elasticity. It is assumed that it does not yield or fracture. Intrinsic to this is the assumption that hydrogen does not attack the diamond.

It was assumed that sticking conditions hold at the diamond-gasket interface. It is assumed that the yield strength of hydrogen is zero, i.e., that it behaves as a fluid with an equation of state as determined by Loubeyre *et al* [14]. This gives a lower bound on the pressure for a given sample radius at pressure. It is assumed that the yield strength of the gasket at a given radius



**Figure 2.** Hydrogen sample radius versus pressure. The initial radius  $r_s^0 = 10 \mu\text{m}$ . The initial bevel was  $8^\circ$ ;  $\sigma_{00} = 8 \text{ GPa}$ . The ideal plasticity limit is 224 GPa.

does not vary with angle of rotation (the real world is not so kind), i.e., that the plastic behaviour is homogeneous. A pressure versus sample-radius plot is shown in figure 2 for  $r_s^0/r_t = 1.0$ .

Note that at low pressure,  $r_s$  decreases as  $P$  increases, but then begins to increase, and then increases further until an instability is reached (at  $P \approx 224 \text{ GPa}$ ) and the pressure then decreases. This is called the ideal plastic instability limit. At the Carnegie Institute, Mao and Hemley used  $r_s^0/r_t = 0.9$  [16].

For  $r_s^0/r_t = 0.9$ , the ideal plastic instability limit is 242 GPa. Even with hydrogen having some shear strength at that pressure and a high coefficient of friction, it is doubtful that a pressure above 250 GPa could be reached.

This explains why their maximum pressures on hydrogen are limited to 230 GPa, [17]. It also explains why they often see the loss of hydrogen although a near-circular gasket hole is still present [16]. Were they to use  $r_s^0/r_t = 0.5$  this problem would disappear (until very much higher pressures were reached) as we see later.

Actually many other (nonideal) things could go wrong. Hydrogen could dissolve in the diamond and migrate to the point where the stresses  $\sigma_\pi$  and  $\sigma_{rr}$  along the centre axes both become tensile, and lead to fracture of the diamond [10]. Hydrogen could unpin dislocations leading to yielding of the diamond. The diamond tip could be slightly tilted leading to sideward migration of the sample hole. The yield strength of these gaskets near to  $r_s$  could vary with  $\theta$ , resulting in an instability occurring and very abnormal radial growth of the hole occurring near a specific  $\theta$  where the yield stress is a minimum. All of these problems can occur, which is why so far the maximum pressure obtained on  $\text{H}_2$  is only 342 GPa [15]; this was obtained with  $r_s^0/r_t = 0.5$ .

When  $r_s^0/r_t = 0.5$ , calculations shown that  $r_s$  decreases at first and then increases, reaching the ideal plasticity limit of 673 GPa. Suppose the situation is nonideal; suppose that along the way before 673 GPa is reached hydrogen diffuses into the diamond either by bulk diffusion or by dislocation pipe diffusion. The mass of hydrogen in the sample hole decreases. The result is that the pressure drops and  $r_s$  decreases. It must be emphasized that  $r_s$  decreases. This has been directly observed in a case in which it occurred on a reasonable timescale. The pressure was 220 GPa and the nut was tightened. Then the next x-ray measurements showed a surprising decrease in pressure. On optical examination it appeared that  $r_s$  had decreased slightly (it had been increasing). Careful examination showed a tiny crack appearing 20 mm below the surface on one of the diamonds. The nut was tightened (so as to increase the compression). The pressure was lower,  $r_s$  had subsequently decreased and the crack size grew. This behaviour was

followed for several cycles. With an  $r_s^0/r_t$  ratio of 0.5 and  $P$  substantially less than the ideal plasticity limit (673 GPa in this case), and with the hole remaining approximately circular, the hole closes when the hydrogen sample is lost. Thus in our 342 GPa experiments (at about half the ideal plastic instability limit),  $H_2$  was definitely present or the hole would have closed [15].

When  $H_2$  is at pressures of 342 GPa or higher it is elastically stiff and the shear stress will be large [18], so optical experiments such as Raman studies will fail to show a signal because of severe line broadening.

In order to reach pressures needed for metallization of hydrogen, perhaps 600 GPa, [19] it is necessary to:

- (1) have a small  $r_s^0/r_t$  ratio;
- (2) isolate the diamond from the hydrogen;
- (3) decrease  $r_t$  from the current value, 10–5  $\mu\text{m}$  and possibly even smaller [4];
- (4) assure homogeneity of the yield stress of the gasket material perhaps by using gaskets produced from one-nanometre tungsten powder.

Fulfilling all of these requirements may be arduous.

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