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Modelling Strength and Ductility of Nanocrystalline Metallic Materials

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Abstract The effect of grain refinement on the strength and ductility of metallic materials is investigated. A model in which a single phase material is considered as an effectively two-phase one is discussed. A distinctive feature of the model is that grain boundaries are treated as a separate phase deforming by a diffusion mechanism. Deformation of the grain interior phase is assumed to be carried by two concurrent mechanisms: dislocation glide and mass transfer by diffusion. The model was exemplified by simulating uniaxial tensile deformation of Cu down to the nanometer grain size. The results confirm the observed strain hardening behaviour and a trend for reduction of ductility with decreasing grain size at room temperature.

Keywords : Nanocrystalline materials, Strength, Ductility, Modeling

1. Introduction

It is generally accepted that grain refinement should lead to enhancement of strength metallic materials. According to the classic Hall-Petch relation, a square root dependence of the yield strength should be observed. However, it is clear that there must be a limit to an increase of strength with decreasing grain size. As the diffusion length for vacancy diffusion is reduced as a result of grain refinement, diffusion controlled mass transport across the grains and along grain boundaries gains on significance and may eventually prevail over dislocation glide controlled plastic flow. Besides, the very mechanism of dislocation glide may become non-operational when the grain size drops below a certain critical level, perhaps around 10 nm.[1] Then, an *inverse* dependence of stress on a certain power of the average grain size d will hold, i.e. $\sigma \propto 1/d^2$ or $\sigma \propto 1/d^3$ for diffusion through the bulk of the grain (Nabarro-Herring mechanism[2]) or along grain boundaries (Coble mechanism[2], respectively). There is some evidence in the literature[3,4] that an inverse dependence, or at least a pronounced departure from the Hall-Petch behaviour, is observed in the small grain size range.

Ductility usually deteriorates with increase of strength, but expectations have been expressed in literature[5,6] that for the grain size in the sub micrometer range, enhancement of strength without loss, or even with an increase, of ductility can be achieved. Particularly with materials that have undergone severe plastic deformation, e.g. by equal channel angular extrusion,[7] such expectations were raised in literature.[8]

In the present paper, a model is shown[9,10] to provide an adequate description of the grain size dependence of strength. A distinctive feature of the model is that grain boundaries are treated as a separate phase that is assumed to deform by a diffusion mechanism. The other 'phase', *viz.* the grain interiors, is considered to deform by a combination of two mechanisms: dislocation glide and diffusion transport. Combined with a rule of mixtures, the constitutive equations for the mechanisms considered provide a simple description of the deformation behaviour. This constitutive model presented will be applied to the case of Cu and compared with the literature data on the grain size dependence of the offset yield stress $\sigma_{0.2}$ and ductility as characterised by the strain at the onset of necking.

2. The constitutive model

The constitutive description of the deformation behaviour of a single-phase material proposed in Refs. [1, 9-11] makes use of the idea[12] that the grain boundaries can be treated as a separate phase. In the previous work[11] it was described as a viscous pseudo-amorphous phase. In a more recent version of the model,[9] plastic flow of this phase was assumed to be controlled by diffusion. The corresponding plastic strain rate is given by

$$\dot{\varepsilon}_{GB} = A\frac{\Omega\sigma_{GB}}{kT}\frac{D_{GB}}{d^2}. \quad (1)$$

Here σ_{GB} is the stress acting in the grain boundary (GB) phase, Ω the atomic volume, T the absolute temperature, k the Boltzmann constant and D_{GB} the self-diffusion coefficient for grain boundary diffusion. The numerical coefficient A was taken to be equal to 2.

Plastic flow of the other phase, *viz.* the crystalline grain interiors, is considered to be carried by two mechanisms that operate in parallel: the dislocation glide mechanism (contributing the plastic strain rate $\dot{\varepsilon}_{disl}$) and the diffusion controlled mechanism. In principle, both bulk and grain boundary diffusion need to be considered. However, for the particular case to be considered below, i.e. deformation of Cu at room temperature, the bulk diffusion contribution can be neglected.[9] The grain boundary diffusion (Coble) mechanism is thus the one operating predominantly. Accordingly, the plastic strain rate for the grain interior (GI) phase can be written as

$$\dot{\varepsilon}_{GI} = \dot{\varepsilon}_{disl}(\sigma_{GI}) + \dot{\varepsilon}_{Coble} \quad (2)$$

where

$$\dot{\varepsilon}_{Coble} = 14\pi\frac{\Omega\sigma_{GI}}{kT}\cdot\frac{w}{d}\cdot\frac{D_{GB}}{d^2}. \quad (3)$$

Here σ_{GI} is the stress acting in the grain interiors and w is the grain boundary width. We note that the latter contribution to the strain rate is similar to the expression for the plastic strain rate of the grain boundary phase, Eq. (1).

The dislocation glide contribution to the plastic strain rate of the grain interior phase is described in terms of a model[13] based on the dislocation density evolution. The total dislocation density is considered as an internal variable whose evolution is affected by the grain size. The relation between the plastic strain rate and stress is given by

$$\dot{\varepsilon}_{disl} = \dot{\varepsilon}_o\left(\frac{\sigma_{GI}}{\sigma_o}\right)^{m_i} Z^{-m_i/2}\Theta(d-d_c) \quad (4)$$

where Z denotes the non-dimensional dislocation density and the parameters $\dot{\varepsilon}_o$, σ_o and m_i can be considered constant for a given temperature. The parameter m_i is temperature dependent, and can be written[13] as $m_i = A/T$, where A = const. The values of the parameters for Cu are known, cf. Ref. [14]. The Heviside step-function expresses the fact that for d below some critical value d_c, the dislocation contribution to plastic strain rate, $\dot{\varepsilon}_{disl}$ drops to zero, the dislocation mechanism becoming non-operative. Following Ref. [1], $d_c = 8$ nm was taken. (According to the definition of the Heviside step-function, for $d > d_c$, $\Theta(d - d_c) = 1$ holds.)

The evolution equation for the dislocation density reads

$$\frac{dZ}{dt} = \dot{\varepsilon}_{disl}\left[C + C_1\sqrt{Z} - C_2\left(\frac{\dot{\varepsilon}_{disl}}{\dot{\varepsilon}_o}\right)^{-1/n} Z\right] \quad (5)$$

Here t denotes the time, C, C_1, C_2 and $\dot{\varepsilon}_o$ are constants, while n is temperature dependent: $n = B/T$, B = const. Thus, the dislocation glide mechanism involves two rate sensitivities: the instantaneous strain rate sensitivity of the *flow stress* represented by $1/m_i$ and the strain rate sensitivity of *strain hardening* that is associated with the dynamic recovery term in Eq. (5) and is given by $1/n$. Grain size effects, which are of main concern here, enter through the term C that is inversely proportional to d.[13]

It is further assumed that the strain in the grain interior and the grain boundary phase is the same (and equal to the macroscopic strain). Henceforth, the subscripts GI and GB in the respective strain rates will be omitted. The stress is determined using a rule of mixtures as a weighted sum of the individual contributions of the two phases:

$$\sigma = f\sigma_{GB} + (1-f)\sigma_{GI}, \quad (6)$$

where f is the volume fraction of the grain boundary phase determined by

$$1 - f = (1 - w/d)^3. \quad (7)$$

(Here cube shaped grains were assumed for simplicity.)

The set of equations (1)-(7) furnish a full constitutive description for a single-phase material treated as a two-phase one. It has been applied for the case of Cu[9-11] for which the parameters pertaining to the dislocation mechanism are known from previous work[14] and those pertaining to the self-diffusion through grain boundaries can be taken from literature [2,15]. It should be noted that data for nanocrystalline copper[15] show a significantly higher grain boundary diffusivity than that in coarse grained Cu.[2] In the present calculations, the former values were chosen as the relevant ones for the range where the diffusion mechanism of plasticity

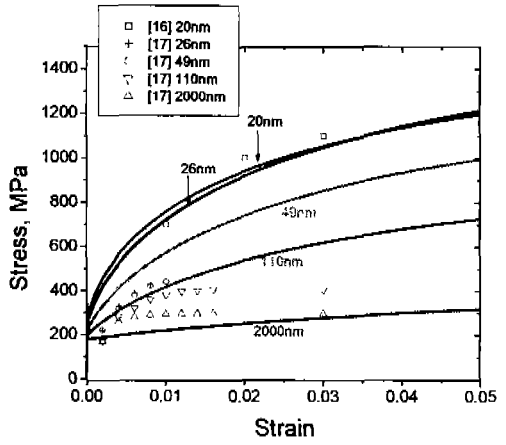

Fig. 1. Calculated and experimental stress-strain curves for Cu at the strain rate of 10^{-3}/s.

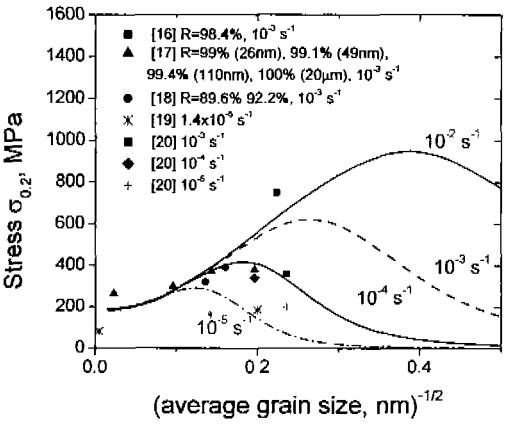

Fig. 3. Average grain size dependence of $\sigma_{0.2}$ for fine grain Cu at strain rates of 10^{-2}, 10^{-3}, 10^{-4}, and 10^{-5} s^{-1} calculated using a log-normal distribution of the grain volume. Also shown are the experimental data points, cf. Fig. 2.

prevails.

As an example, the stress *vs.* strain curves for tensile deformation with constant strain rate are presented in Fig. 1. The curves show a good agreement with experimental data from literature. It should be stressed that this agreement was achieved without use of any adjustable parameters.

The dependence of the offset yield stress $\sigma_{0.2}$ on the grain size calculated using this model is plotted in the Hall-Petch diagram, Fig. 2. It is seen that a Hall-Petch-like behaviour in the range of large grain sizes breaks down for d below about 50 nm, and an 'inverse' Hall-Petch behaviour is predicted for small values of d and $\dot{\varepsilon}$. The numerical correspondence between the calculated curves and the experimental results is rather poor, but the main trends predicted seem to be confirmed by experiments. It should be mentioned in this connection that the bulk of experimental data were obtained on specimens containing residual porosity cause by the fabrication technique used. Accordingly, higher values of yield strength should be expected for fully dense material, which would improve the agreement between modelling and experiment.

Even with the inferior quality materials tested, a better agreement is achieved if grain size distribution is taken into account. The $\sigma_{0.2}$ vs. $d^{-1/2}$ curves obtained using a log-normal grain volume distribution[10] are shown in Fig. 3. It is evident that an improved correspondence between calculation results and experiment is achieved in this case. This example illustrates the

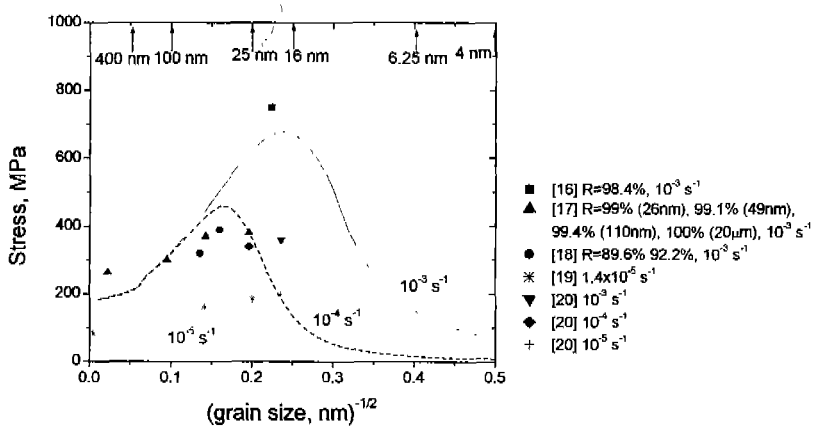

Fig. 2. Calculated grain size dependence of $\sigma_{0.2}$ for nanocrystalline Cu for strain rates of 10^{-3}, 10^{-4} and 10^{-5} s^{-1}. Symbols correspond to experimental data from literature. The relative material density R is indicated where available.

importance of inclusion of grain size distribution in modelling exercises. It can be seen that grain size distribution leads to lower and broader stress peaks. It should be noted that the strength in the coarse grain size range is altered slightly, whereas the change in the fine grain size range, e.g. for $d < 100$ nm, is fairly pronounced. This is attributed to the fact that for log-normal grain volume distribution, the volume fraction of large grains is much larger than that of fine grains (although the number of large grains may not be greater than that of fine grains).

3. Tensile ductility

The model presented in the previous section can also be used to determine the grain size dependence of the tensile ductility. Koch and Malow[21] who compiled ductility data demonstrated that ultra fine grained single-phase materials exhibit little ductility in tension. For elemental metals that are ductile when coarse grained, the data show a clear trend for decreasing tensile ductility with decreasing grain size, cf. Fig. 4. A similar behaviour of ductility of Al alloys with grain size is reported in a recent publication.[22]

In a recent communication,[27] the effect of grain size on ductility was considered by analysing the condition for the onset of necking. Harts necking condition,[28]

$$\theta \leq \sigma(1 - 1/m), \tag{8}$$

was used to that end. Here the strain hardening coefficient and the effective strain rate sensitivity parameter are defined as $\theta = (\partial\sigma/\partial\varepsilon)_{\dot{\varepsilon}}$ and $1/m = (\partial log\sigma/\partial log\dot{\varepsilon})_{\varepsilon}$, respectively. It can easily be seen from this criterion that for extremely fine grained materials, when plasticity is carried exclusively by the diffusion mecha-nisms and m tends to unity, tensile deformation will always be stable, strain-to-necking tending to infinity, suggesting the occurrence of super-plasticity. That is to say, the material will never fail by necking, its ductility being limited by other failure mechanisms. In a real case, even for a very small grain size, the fulfilment of the necking condition, i.e. the equality in Eq. (8), will depend on the interplay of m, σ and θ.

Numerical simulations of a strain rate jump test can be used to determine the dependence of the instantaneous strain rate sensitivity $1/m$ on the grain size (Fig. 6). These numerical experiments showed that the strain dependence of the rate sensitivity parameter $1/m$ is fairly weak. The results presented in Fig. 6 refer to the proof strain of 0.2%. Substituting the calculated values

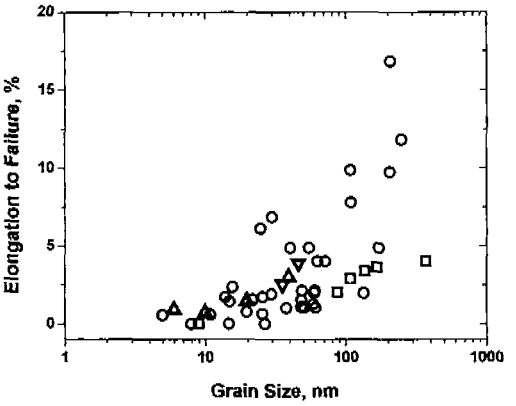

Fig. 4. Tensile ductility of single-phase materials as a function of grain size. All data are for room tem-perature (after Koch and Malow[21]). Symbols: □ Al/Al alloy, ○ Cu, △ Ni, ▽ Pd.

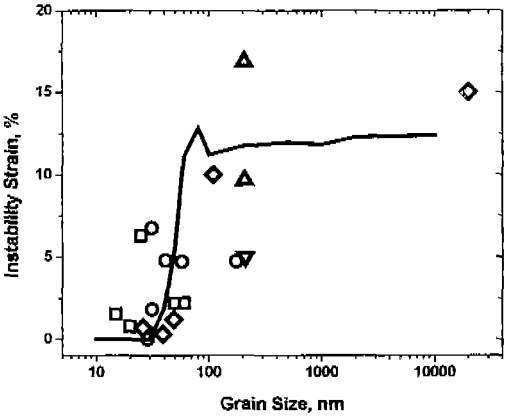

Fig. 5. Room temperature ductility of Cu as a function of the grain size: literature data (symbols) vs. model prediction (solid line). Symbols: □ Ref. [23], ○ Ref. [24], △ Ref. [25], ◇ Ref. [17], ▽ Ref. [26].

of σ, θ and $1/m$ in Hart's condition, the grain size dependence of strain-to-necking was determined. This dependence is shown as a solid line in Fig. 5.

The results presented in Fig. 5 demonstrate that the experimentally observed reduction of tensile ductility with grain refinement is in close quantitative agreement with the model. The loss of ductility with grain refinement found for room temperature deformation of copper is not a generic trend with all materials at all temperatures. It rather depends on the particular values of parameters determining the rate sensitivity of stress, as well as the level of stress and the rate of its variation with strain. The effect of temperature on the grain size

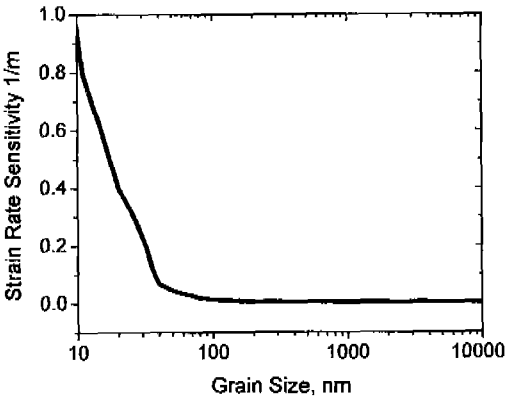

Fig. 6. Grain size dependence of the strain rate sensitivity parameter 1/m.

dependence of strain-to-necking will be discussed in a forthcoming publication.

4. Conclusion

A relatively simple model based on subdivision of a single phase material in two separate phases deforming by different mechanisms was presented. The grain boundary phase plays a prominent role in this model approach. Grain size effects were introduced both via the dislocation glide mechanism and through the diffusion mechanisms providing mass transfer via grain boundaries. A good agreement between the calculated deformation behaviour and experiment was found. The quality of the above predictions with regard to strength and strain hardening behaviour testify the adequacy of the model. On a practical level, the model works very well and can be used as a convenient tool for simulating the deformation behaviour of nanocrystalline metallic materials.

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