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ON THE WORK HARDENING OF FIBER REINFORCED COPPER

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Introduction

The prediction and optimization of metal matrix composite mechanical behavior necessitates an understanding of the influence exerted by the reinforcement on matrix plastic deformation. This influence can be substantial, as was first eloquently shown in 1969 by Kelly and Lilholt (1). In this work, composites of continuous 10 and 20 μm diameter tungsten fiber reinforced copper composites were produced and tested in tension along the fiber direction. This system was chosen for its simplicity, both in structure and in processing: copper and tungsten feature no mutual solubility nor intermetallic phases (2), and copper wets tungsten well. The composites were fabricated by spontaneous infiltration of molten copper into packed bundles of parallel fibers held within cylindrical molds, followed by directional solidification to ensure proper feeding of metal solidification shrinkage. Resulting composites were free of pores, and their matrix was found to be essentially monocrystalline. In processing the composites, the fiber volume fraction V_f was carefully measured by counting the number of fibers in each preform, checking that the fiber diameter was in good agreement (within 1%) with the nominal fiber diameter (3). V_f values thus determined were also verified using measurements of composite density, to find good agreement; quoted V_f values can thus be deemed reliable.

Fiber stress-strain curves were measured using fibers subjected to the same thermal cycle as the composites. With these composites, the rule of mixtures is valid within close bounds (1,4,5). Therefore, as long as the fibers remain elastic, and beyond fiber yield if residual stresses are ignored, the average *in-situ* matrix flow stress σ_m could be calculated from the composite flow stress, σ_c, and the fiber flow stress, σ_f, according to:

$$\sigma_m = \frac{\sigma_c - V_f \sigma_f}{1 - V_f}. \quad (1)$$

The composites were found to exhibit three stages on their stress-strain curves: (i) an elastic range (Stage I), of slope in most cases within the range of variation of the predicted composite elastic modulus, $E_f V_f + E_m (1 - V_f)$, variation in the predicted composite modulus being due to the considerable anisotropy in copper single crystal elastic moduli (the anisotropy ratio A equals 3.21 for

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Cu (6)), (ii) an intermediate range (Stage II), in which the composite featured permanent plastic deformation, of lower but roughly constant slope, and (iii) a range of lower still and often decreasing slope (Stage III), observed at strains roughly above the yield strain of the independently tested fibers.

These experiments resulted in the striking observation that measured *in-situ* matrix work hardening rates during Stage II are very high, being within an order of magnitude of the matrix modulus range of variation. For comparison, the maximum rates of work hardening measured in pure metals are of the order of the shear modulus divided by about 20 (7). Two additional and important observations were that during Stage II, the measured *in-situ* rates of matrix work hardening (i) increased with increasing V_f roughly proportionally to $V_f(1 - V_f)^{-1}$, and (ii) increased somewhat as the fiber diameter decreased at constant V_f from 20 to 10 μm.

Several explanations have been proposed to interpret these data. Kelly and Lilholt, after discounting individual dislocation pile-up models on quantitative grounds, proposed that their data are explained if one postulates the existence of a region, about 3 μm wide, of matrix surrounding each fiber, this region remaining elastic until the fibers yield (1). Subsequent interpretations include a model by Neumann and Haasen, which assumes the formation of elastically interacting parallel dislocation pile-ups (8), and a "source-shortening" model by Brown and Clarke (9), which quantifies the resistance to dislocation glide offered by the internal stresses from Orowan loops left around fibres. Detailed discussions of the many models that have been proposed to explain the increased rate of matrix work hardening found in fiber reinforced composites can be found in Refs. (10,11). It is concluded that a so called "elastic friction" effect similar to source-shortening, but caused by fluctuating internal stresses due to the phase modulus difference, rather than Orowan loops, must be invoked to account for the strong apparent matrix work hardening rate at intermediate to high fibre volume fractions.

The aim of the present note is to propose an alternative explanation for the stage II matrix hardening rates measured in infiltrated copper-tungsten composites. The explanation is close to the original interpretation, but proposes an observable physical basis for the occurrence of plastically non-yielding matrix regions in Stage II.

A Model Based on Fiber Clustering

The present model is based on two observations. The first is that, when a composite is produced by infiltrating loosely packed fiber bundles with a liquid matrix, there is a natural tendency for the fibers to congregate into clusters (just as partly wet straight hair tends, in air, to congregate into separate strands). The physical reason for such clustering is the formation, when two fluid phases (vapor phase and liquid matrix in composite infiltration) coexist in the fiber preform, of menisci of the better wetting phase between neighboring fibers, Fig. 1. These menisci exert forces on the fibers, which tend to pull them together. Resulting forces correspond to pressures on the order of $P = \gamma/r_f$, γ being the surface tension of the liquid matrix and r_f the fiber radius. Metal surface tensions γ being on the order of 1 J $\cdot$ m^{-2}, with r_f near 10 μm, corresponding pressures are near one atmosphere: this is certainly sufficient to pull together fibers if these are not tightly compressed in their mold.

With fine fibers, near 10 or 20 μm in diameter, little control can therefore be exerted on the local fiber volume fraction unless special techniques are used (such as the "hybridization" process (12)). If such techniques are not used, when parallel-fiber bundles are packed to a volume fraction V_f below the value $V_{f,max}$ to which fibers are compressed under a pressure on the order of γ/r_f, the resulting composite features regions of fibers clustered to a volume fraction near $V_{f,max}$, separated by channels of approximately fiber-free matrix.

This problem was recognized by Kelly and Lilholt, who wound copper wires 60 μm in diameter between the tungsten fibers prior to infiltration when preparing low volume fraction fiber preforms (3).

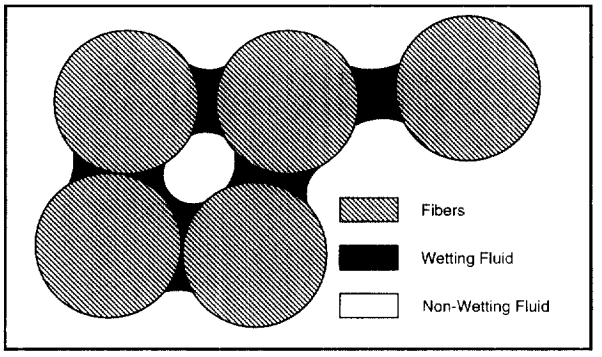

Figure 1. Schematic depiction of menisci of a wetting fluid between fibers in the presence of a non-wetting fluid, illustrating how capillary forces pull fibers together during intermediate stages of the infiltration process (in infiltration, one phase is the liquid matrix, the other air, the nature of the phases depending on the wetting angle of the molten matrix on the reinforcement in air).

The copper wires prevented the fibers from clustering into very large close-packed bundles in the composites produced; however, since the copper wires were molten at the temperature of infiltration, these did not prevent lateral motion of the fibers during infiltration—in fact, the molten copper wires must have caused local fiber clustering before infiltration. The infiltrated composites should therefore have featured fiber segregation into several small close-packed bundles, distributed over the sample cross section. Such fiber clustering is indeed seen in Fig. 1 of Ref. (1), and in Fig. 18 of Ref. (3).

The second observation we use to interpret the data relates to a composite sample, containing 48 vol.% 20 μm fibers, which is reported in Ref. (3). In testing, this sample remained in Stage I up to a strain of 0.15%; data were off-scale thereafter on the experimental stress-strain curve, which is why this sample was excluded from the list of data in Ref. (1). For all other samples, of $V_f < 48\%$ and reported in Ref. (1), initial yield, causing the observed transition from Stage I to Stage II, occurred at a much lower strain, near 0.02%.

In the composite containing 48 vol.% fibers, therefore, the matrix yield stress is everywhere higher than about 350 MPa, a rather high value for pure copper. The physical origin of such a high matrix yield stress in Cu-W composites has not been specifically identified; however, it is highly plausible that is caused by differential contraction of the matrix around the fibers during cool-down of the composite from processing temperatures: the coefficients of thermal expansions of copper and tungsten are quite different, at $17 \cdot 10^{-6}$ and $4.5 \cdot 10^{-6}$ K^{-1}, respectively (13). Such differences in thermal contraction between a metallic matrix and a reinforcement in a composite are well known to cause significant increases in dislocation density within the matrix. For example, fully packed (to V_f of 45 to 50 vol.%) 20 μm alumina fibers in pure aluminum cause, during cooldown from the matrix melting point, the formation of matrix substructures similar to those found in aluminum deformed to very high strains (14). Since pure copper can indeed possess a flow stress as high as 300 MPa at room temperature after large strain deformation (7), it is quite reasonable that dislocation punching by the fibers into the matrix during composite cool down would be responsible for the observed lack of yielding at a strain of 0.15% in a Cu-W composite containing 48 vol.% fibers.

It is known that, in practice, fibers 20 to 10 μm in diameter can be packed in a mold to a maximum volume fraction around 45 to 55% before significant fiber breakage (for ceramic fibers), or deformation (for metal wires), occurs at fiber contact points (*e.g.*, Ref. (14)). It is additionally documented in Ref. (3) that the maximum fiber volume fraction practically attainable in the experiments was indeed about 50%. The composite produced with 48% wires 20 μm in diameter must, therefore, have been relatively free of wide fiber-free copper channels. We therefore adopt the value 48% as an estimate of the volume

fraction, $V_{f,max}$, to which their tungsten fibers cluster under the action of capillary forces during infiltration.

When V_f is lower than $V_{f,max}$, because capillary forces pull fibers together during infiltration, the infiltrated composite microstructure is better (albeit still approximately) described as a mixture of fiber-free copper regions adjoining regions of Cu-W composite, containing clustered tungsten fibers of $V_f = V_{f,max}$. Applying the rule of mixtures to the resulting composite of unreinforced copper and close-packed tungsten fiber reinforced copper composite, the composite stress σ_c is then given by:

$$\sigma_c = V_c\sigma_{c,max} + (1 - V_c)\sigma_{Cu} \tag{2}$$

where V_c is the volume fraction of composite made of clustered fibers and copper located between close-packed fibers, for which $V_f = V_{f,max}$, and which flows at stress $\sigma_{c,max}$. Since $V_c = V_f/V_{f,max}$, at strains less than 0.15%, the stress-strain relation of the Cu-W composites should approximately be given by:

$$\sigma_c = V_f E_f \varepsilon + V_f \frac{1 - V_{f,max}}{V_{f,max}} E_{Cu}\varepsilon + \left(1 - \frac{V_f}{V_{f,max}}\right)\sigma_{Cu} \tag{3}$$

where σ_{Cu} is the matrix flow stress at strain ε in the unreinforced condition, characteristic of matrix areas devoid of fibers.

Wider unreinforced copper channels should contain far fewer dislocations than copper channels situated between close-packed fibers. It is therefore expected that their yield strain would be much lower than that of copper located between clustered fibers: with $V_f < V_{f,max}$, then, composites must yield when plastic deformation begins within softer fiber-free copper channels. This explains why, with $V_f < 48\%$, the transition from Stage I to Stage II was observed at a strain near 0.02%, typical of unhardened copper and far lower than the matrix yield strain when V_f equalled 48%. From Eq. (3), the derived apparent *in-situ* matrix stress σ'_m and the derived apparent *in-situ* rate of matrix work hardening are then predicted, respectively, to equal in Stage II:

$$(\sigma'_m)_{(\text{Region II})} = \frac{V_f(1 - V_{f,max})}{V_{f,max}(1 - V_f)} E_{Cu}\varepsilon + \frac{1}{(1 - V_f)}\left(1 - \frac{V_f}{V_{f,max}}\right)\sigma_{Cu} \tag{4}$$

and

$$\left(\frac{d\sigma'_m}{d\varepsilon}\right)_{(\text{Region II})} = \frac{V_f(1 - V_{f,max})}{V_{f,max}(1 - V_f)} E_{Cu} + \frac{1}{(1 - V_f)}\left(1 - \frac{V_f}{V_{f,max}}\right)\frac{d\sigma_{Cu}}{d\varepsilon} \tag{5}$$

where $(d\sigma_{Cu}/d\varepsilon)$ is the rate of matrix work hardening in the unreinforced condition, roughly equal to 1 GPa from the slope of the stress-strain curves of copper single crystals processed identically to the composites (1).

Comparison with Data and Discussion

Several difficulties exist in comparing data with theory. One of these stems from the large dependence, in copper, of the tensile elastic modulus with crystal orientation. In the infiltrated Cu-W composites, the matrix was reported to be single-crystalline, with the consequence that E_{Cu} in each composite varied widely; this is apparent in matrix moduli measured in Stage I of composite deformation (1,3). We have therefore chosen to use these reported values of E_{Cu} when computing from Eq. (5) the predicted apparent matrix rate of work hardening for each composite. This choice seems the most logical: however, should any of the composites not be single-crystalline, significant error would result.

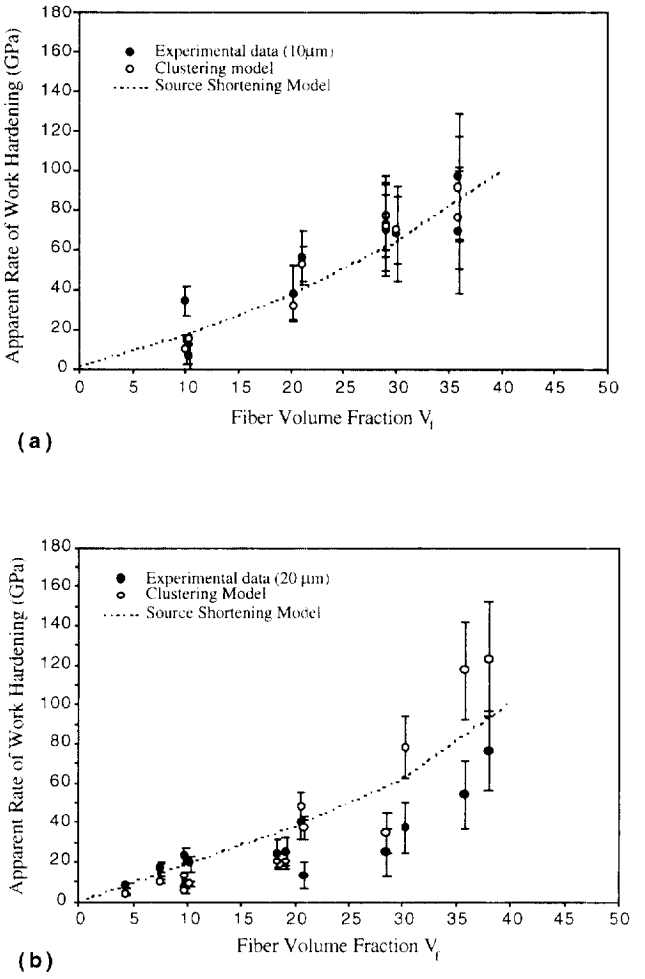

Figure 2. Comparison between observed and predicted apparent *in-situ* matrix work hardening rate in composites of copper reinforced with (a) 10 μm and (b) 20 μm tungsten fibers.

particularly at low V_f. Additionally, it is stressed that experimental error in measured *in-situ* matrix apparent moduli or work hardening rates is very significant, in particular because of experimental error in (E_f V_f). This, too, was carefully considered by Kelly and Lilholt, who reported experimental error estimations for their data, these estimations being based on uncertainty in E_f (1). As an illustration of the level of uncertainty involved, note that the measured Stage II matrix *in-situ* rates of work hardening differ by as much as half an order of magnitude for the three samples containing 10 vol.% 10 μm fibers, Fig. 2.

A comparison of measured and derived apparent rates of matrix work hardening in Stage II are given in Fig. 2. Reported experimental error is from Ref. (1), while error in the predictions from Eq. (5) is due to error in the value of E_{Cu}, also reported in Ref. (1). It is seen that, although the agreement between derived and measured rates of matrix work hardening is far from perfect, the level of agreement is quite acceptable given the considerable uncertainties involved in the comparison, for example in the assumed value for $V_{f,max}$, and in the fact that fibers are not perfectly clustered, as assumed in deriving Eq. (5). Furthermore, such variations in fiber packing from sample to sample would, in the framework of the

present interpretation, explain the significant variations in measured apparent matrix rate of work hardening that are observed across similar samples.

The present explanation for the data also provides a rationalization for the observation that apparent matrix work hardening rates seem to increase somewhat as the fiber diameter decreases from 20 to 10 μm. Finer fibers tend to be more difficult to align, such that these would tend to form clusters of lower volume fraction fiber $V_{f,max}$. Finer fibers also cause, in principle at least, the emission of a greater length of dislocations per unit matrix volume during composite cool down; hence, it is also likely that fiber clusters of somewhat lower $V_{f,max}$ would, with 10 μm fibers, contain a sufficient density of dislocations to delay matrix yield significantly past a strain of 0.02%. From Eq. (5), a decrease in $V_{f,max}$ would indeed, all else being equal, cause an increase in $(d\sigma'_m/d\varepsilon)_{Region\ II}$. Also, since much thicker fibers, 100 μm or more in diameter, cause far less dislocation emission per unit volume of matrix during composite cool-down from processing temperatures, no dramatic increase in apparent matrix work hardening rate is expected in Cu-W composites produced with large W fibers: this is substantiated by experiment (e.g., (11,15)).

Generally, the earlier models have taken as a starting point that Kelly and Lilholt's data together with experimental data from other studies of W-Cu fiber composites, indicate a strong effect of the average interfiber spacing on the rate of matrix hardening during plastic deformation, narrow interfiber spacings causing high apparent matrix hardening rates. Fig. 2 provides a comparison between predictions from the present model, and from the earlier source-shortening/elastic friction model (10,11). It is seen that both models provide quite acceptable agreement, the clustering model overestimating more strongly than the source-shortening/elastic friction model the apparent matrix *in-situ* rate of work hardening for 20 μm fibers.

The two models are thus comparable in terms of their overall agreement with experiment, and also in their basic premise, since both invoke a dislocation mechanism for the occurrence of non-yielding regions of matrix. Both models therefore capture the dependence of the apparent matrix *in-situ* work hardening rate on fiber volume fraction. The models differ somewhat in regard to the observed dependence on fiber diameter. In the source-shortening/elastic friction model the dependence is explained entirely as a scale-dependent stress relaxation rate, which subtracts from the apparent matrix hardening rate. The exact nature of the stress relaxation process has been discussed by several investigators, e.g., (16–18), using in part the observation (11) that for 100 μm fibers relaxation occurs even at a temperature as low as 77K, where diffusion is very slow in copper, which suggests that relaxation involves dislocation glide rather than diffusion and climb. In the clustering model a possible source of scale dependence can be found in the expected increase in punched dislocation density with decreasing fiber diameter, and in the dependence on fiber diameter of the extent of clustering, as represented in this one-parameter model by $V_{f,max}$. Thus, the clustering model adds the new possibility that stress relaxation is controlled by the scale-dependent density of dislocations generated inside the fibre clusters during cool-down from the infiltration temperature. Indeed, static relaxation of internal stresses generated by composite cool-down has been observed in fiber reinforced aluminium (19).

Summary

We offer a simple interpretation of the observation of abnormally high rates of matrix work hardening observed during Stage II deformation of infiltrated Cu-W composites: we show that the enhanced matrix hardening rates could result from fiber clustering during infiltration followed by dislocation hardening of the matrix inside clusters during cool-down from the processing temperatures. Our explanation is similar in nature to that advocated initially by Kelly and Lilholt in their paper, namely that a portion of the matrix stays elastic throughout stage II of tensile deformation. What we provide

is a physical explanation for the formation of such an elastic portion of the matrix, based on the physics of composite processing, on the observed clustering of fibres in the microstructure of the composites, and on data from an additional sample reported in the original thesis (3).

The matrix hardening predicted by the present model is in good agreement with the experimental data. It is interesting to note that the predicted matrix hardening rates are comparable with those predicted by the earlier source-shortening/elastic friction model, although the two models are based on distinctly different dislocation mechanisms. For high volume fractions of 20 μm diameter fibres the present model overestimates the apparent matrix work hardening rate somewhat more than does the earlier model; in both models the overestimate is consistent with the operation of a scale-dependent stress relaxation process.

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