Analysis of the Grain Size Dependence of the Yield Stress in Copper-Aluminum and Copper-Nickel Alloys

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Grain size dependence of the yield stress in polycrystalline copper-aluminum and copper-nickel alloys is measured at 77, 194 and 293 °K. It is found that the Hall-Petch relation is valid for these materials. The Petch slope $k_y$ is nearly independent of the temperature, but increases with increasing solute concentration. In copper-aluminum alloys the increase in $k_y$ is nearly proportional to the square root of the solute concentration, while in copper-nickel alloys it is proportional to the concentration itself. The concentration dependence of $k_y$ in Cu-Al alloys is considerably larger than that in Cu-Ni alloys. None of previously proposed theories on the Petch slope can explain these results. The experimental results will be compared with the calculation in the following paper.

(Received May 7, 1974)

I. Introduction

Grain refinement is a useful method for strengthening polycrystalline metals. Hall(1) and subsequently Petch(2) have established experimentally a simple relation between the lower yield $\sigma_y$ of mild steel and the average grain diameter $d$,

$$ \sigma_y = \sigma_i + k_y d^{-1/2} \tag{1} $$

where $\sigma_i$ and $k_y$ are constants independent of $d$.

Many works have been carried out to examine the validity of eq. (1) for various polycrystalline metals and alloys. As the result of these investigations, eq. (1) has been shown to be valid for such body-centered cubic metals as iron and steel(2)-(11), molybdenum(12) and niobium(13), and for such face-centered cubic metals as copper(14), aluminum(15)(16), silver(17)(18), aluminum 3.5% magnesium alloy(19), copper-zinc alloys(10)(20)(21)(22), copper-aluminum alloys(23)(24), nickel-carbon alloys(25)(26), nickel-cobalt-carbon alloys(26) and iron-nickel alloys(27) and also for such hexagonal close-packed metals as titanium(28), zirconium(29), magnesium(30), and zine(10).

On the other hand, Fleischer(31), opposing to eq. (1), has proposed that the yield stress of copper alloys are inversely proportional to the average grain diameter on the basis of his measurements. Recently Anderson and his co-workers(32) have re-examined the validity of eq. (1) for Armco iron at various temperatures and strain rates, over a wide range of grain sizes. They indicate that the plot of $\sigma_y$ against $d^{-1/2}$ is not linear but is concave toward the $d^{-1/2}$ axis over the range of $d^{-1/2}$ from 7 to 53 cm$^{-1/2}$.

The relation (1) has been discussed in terms of dislocation theory. Hall and Petch have proposed a microscopic model based on the stress concentration caused by a pile-up group of dislocations at a grain boundary. As the stress concentration reaches a critical value, it will generate mobile dislocations in the neighboring grain, and the yielding occurs. This stress concentration has been calculated for a single-ended pile-up of dislocations by Eshelby et al.(33) and for a double-ended pile-up by Leibfried(34) and Head et al.(35) These calculations are extended by Chou et al.(36) and Armstrong et al.(37) to the case of an extremely small grain size down to several hundred angstroms. An excellent review on the calculations of the stress concentration caused by pile-ups, especially by multi-layer pile-ups, is given by Li(38).

In addition to these Li(39) explains the Petch relation from a consideration of dislocation sources at the grain boundary without using the pile-up model.

If the pile-up model is assumed in the case of bcc metals, especially iron and steel, the Petch slope $k_y$ in eq. (1) seems to be reasonably interpreted by considering the break-away of dislocations from their Cottrell atmospheres or the creation of dislocations in a perfect lattice, depending on the impurity concentration and the previous heat treatment of the specimen(40)(41). Meanwhile, in fcc metals and alloys there is no successful interpretation of $k_y$. Though one of the present authors(20) attempted to interpret $k_y$ in $\alpha$-brass by considering the break-away of dislocations from their solute atmospheres, the dislocation density in a recrystallized specimen is so small that there are no dislocations suitable to break-away by the stress concentration. Also we can not ignore that some experiments on fcc alloys contradict eq. (1)(31). Further investigations are, therefore, necessary for fcc metals and alloys from both experimental and theoretical points of view.

The purpose of this paper is to re-examine the validity of the Petch-Hall relation in Cu-Al and Cu-Ni alloys, and also to offer the data to discuss the mechanism of yielding of the polycrystalline fcc alloys.

II. Experimental Methods

The copper-aluminum alloys were melted to compositions of 0, 0.5, 1.0, 5.0, 10.0 and 15.0 at% aluminum, from copper of 99.999% purity and aluminum of 99.999% purity. The copper-nickel alloys were...
melted to composition of 5, 15, 25 wt% nickel, from copper of 99.98% purity and from nickel of 99.8% purity. Each ingot of these alloys of about 1 kg in weight was swaged, rolled and finally cold drawn into wires of 1.7 mm in diameter. The wires were cut into pieces of 55 mm in length and then annealed in vacuum at temperatures between 450 and 1000°C for various time periods. In order to utilize as grips for tensile tests, steel balls of 6.35 mm in diameter were soldered at the both ends of these pieces of wire.

The chemical analyses of aluminum and nickel indicate that the solute concentrations of the final specimens agree with the initially mixed compositions within ±0.2 at%, so that the latter were used as the solute concentrations of specimens hereafter.

Grain size were measured by linear intercept and by area method. The average value was obtained by measuring more than five hundred grains in each specimen. In these measurements twin boundaries are neglected, because twin boundaries do not seem to be strong barriers for the pile-up dislocations as will be discussed in the following paper. A few examples of the relation between the grain size and the annealing temperature are shown in Fig. 1. As seen from this figure, the secondary recrystallization seems to take place at temperatures above 800°C. Tensile tests were performed at 77, 194, 293°C using a hard type testing machine, the specimens being dipped in cryogens. The strain rate was $2.5 \times 10^{-4}$ sec$^{-1}$. The yield stress was defined as the intersection of the tangent to the stress-strain curve at 0.5% plastic strain and the straight line extrapolated from the linear elastic region of the curve, provided that the stress increases steadily with increasing strain. If the stress-strain curve reveals a plateau corresponding to the Lüders strain, the yield stress was defined as the average stress required to propagate the Lüders band.

III. Experimental Results

Figures 2~7 show the yield stress of Cu–Al and Cu–Ni alloys measured at 293, 194 and 77°C as a function of the inverse square root of grain size. In these figures an asterisk indicates the yielding accompanied by the Lüders strain. As easily seen in these figures, the Petch-Hall relation is valid for these alloys.

Figures 8 and 9 show the temperature dependence of the Petch slope $k_p$ in Cu–Al alloys. It is to be noted that the Petch slope in dilute alloys decreases monotonously with increasing temperature, while it reveals...
complicated temperature dependence in alloys containing more than 5 at% aluminum. The situation is similar in Cu–Ni alloys as shown in Fig. 10. Though these temperature dependences of $k_y$ are larger than the error in the experiment, the change in $k_y$ is not significant compared with the magnitude of $k_y$ itself. We may, therefore, regard $k_y$ as nearly independent of the temperature.
As far as we have experienced such a tendency is observed when we are not very careful to handle the annealed specimen. The increase in the yield stress caused by improper handling is significant in soft specimens with low concentrations of solute atoms and having large grain sizes. Then one may obtain a smaller value of \( k_y \) than the true one in low concentration alloys.

As seen in Fig. 1 the secondary recrystallization seems to occur in Cu–10 at.%Al and Cu–15 at.%Al alloys at temperatures above 800°C. It is known that a preferred orientation in polycrystalline metals appears at the stage of the secondary recrystallization. X-ray analysis on specimens containing 10 and 15 at.%Al reveals the \(<111>\) preferred orientation along the specimen axis. The average of the maximum resolved shear stress in \( <110>\{111> \) slip systems over the grains in the specimen having the \( <111>\) preferred orientation is less than that of a randomly oriented specimen by a factor \( 4\sqrt{2}/9 \). Therefore, if the yield stress depends on the average of the maximum resolved shear stress in each grain, the yield stress is strongly affected by the preferred orientation and deviates appreciably from the relation (1). The Petch slope should increase in the region of large grain sizes. Such a tendency must be significant only in high concentration alloys because the preferred orientation was noticeable only in those alloys. The observed yield stresses shown in Figs. 2–7 do not show this tendency. The preferred orientation has little influence on the yield stress, because five independent slip systems must operate in each grain to avoid the misfit of deformation at the grain boundary as already pointed out by Taylor\(^{(43)}\) in 1938. The tensile stress required to cause multiple slip in five slip systems is less dependent on the crystal orientation of each grain than that required for the single slip\(^{(44)}\).

**IV. Discussion**

In order to cause measurable macroscopic plastic strain in a polycrystalline material, slip must propagate through grain boundaries. As mentioned in I the propagation may take place by generating mobile dislocations in the neighboring grain under the stress concentration due to a pile-up of dislocations or by the emission of dislocations from the grain boundary. Possible mechanisms of the propagation of slip through a grain boundary may be described as follows.

1. A dislocation in the neighboring grain operates as a dislocation source under the stress concentration due to the pile-up of dislocations at the grain boundary\(^{(20)(45)}\).

2. A pair of dislocations are created in the neighboring grain under the stress concentration\(^{(40)(45)}\).

3. The leading dislocation of a pile-up passes through the grain boundary leaving a kind of misfit dislocation there\(^{(46)}\).

4. Grain boundaries act as sources of dislocations\(^{(39)}\).
The mechanism (1) does not seem to explain the present experiments, because the locking force of a dislocation cannot be different so significantly as to cause the marked difference in the concentration dependence of \( k_y \) between Cu–Al and Cu–Ni alloys. The dislocation density in a recrystallized \( fcc \) alloys may be less than \( 10^7 \) cm\(^{-2} \). The number of dislocations in a grain with diameter of \( d \) is then less than \( 10^3 \) \( d^2 \) cm\(^{-2} \). Namely a grain with diameter of 10 \( \mu \) contains ten or less dislocations. Then it seems difficult to find out a suitable dislocation which operates as a dislocation source under the stress concentration of the pile-up. The mechanism (1) is, therefore, excluded from the mechanisms applicable to \( fcc \) alloys, in which specimens are usually tested in the as-recrystallized state. Of course it seems to be important in pre-strained or quenched mild steel as demonstrated by Cottrell\(^{(40)} \) and Fisher\(^{(41)} \).

The conventional treatment\(^{(40)} \) of the stress required for the mechanism (2) does not seem to explain the concentration dependence of \( k_y \) because the stress needed to create a pair of dislocations is nearly independent of the solute concentration. It becomes, however, clear that the mechanism (2) is essentially the same as (3), if we take into account the following situation. The stress concentration due to the pile-up is highest at the grain boundary, so that the pair of dislocations may start from the grain boundary. Then one of the dislocations starts from the grain boundary, while the other reacts with the leading dislocation of the pile-up.

The mechanism (4) has been supported by direct observations in silicon-iron alloys\(^{(47)-(48)} \), but the number of dislocations emitted from grain boundaries seems to be limited. Li\(^{(39)} \) assumed that the flow stress at the time of yielding is the stress required to move dislocations in the forest formed by all the dislocations generated from the grain boundary ledges. He obtained a relation same as eq. (1), in which \( k_y \) is given by

\[
k_y = \alpha \mu b m \left( \frac{8p}{\pi} \right)^{1/2}, \tag{2}
\]

where \( \alpha \) is a constant of the order of 0.4 and \( \rho \) is the geometrical factor relating the tensile stress to the shear stress on the operating slip systems. Substituting the observed values of \( k_y \) at room temperature, \( \alpha = 0.4 \) and \( m = 3.1 \)\(^{(43)} \), we obtain the values of \( \rho b \) in eq. (2) as 0.08, 0.52, 1.56 for pure copper, Cu–5 at\%Al and Cu–15 at\%Al, respectively. The distance between the neighboring ledges in the grain boundary should be considerably larger than the atomic distance, so that \( \rho b \) must be much smaller than unity. The magnitudes of \( \rho b \) derived from the observed values of \( k_y \) in high concentration aluminum alloys do not satisfy the condition. The mechanism (4) is, therefore, able to be neglected in the yield point criterion.

The condition of the passage of a dislocation through a grain boundary may be affected by the solute distribution around the grain boundary. A solute atom interacts with the grain boundary by the size and modulus effects. By means of the latter interaction the grain boundary attracts solute atoms when the shear modulus of the alloy decreases with increasing concentration, and repels solute atoms in the reverse case. The former specifies favorable and unfavorable cites of solute atoms in the vicinity of the grain boundary. The shear modulus of Cu–Al alloys decreases with increasing aluminum content, while the reverse is the case in Cu–Ni alloys. Therefore, the stress required to propagate the slip by the mechanism (3) should be strongly affected by solute atoms in Cu–Al alloys, while it would be nearly independent of solute concentrations in Cu–Ni alloys. Observed results reveal the tendency. Further discussions require the quantitative estimation of yield stress assuming the mechanism (3), which will appear in the following paper.

Finally we conclude that the mechanisms (1) and (4) are excluded from the applicable one to \( fcc \) alloys, and that the mechanism (2) is essentially the same as (3), which may explain the concentration dependence of the Petch slope.

V. Conclusions

The relations between yield stress and grain diameter were investigated on Cu–Al alloys containing up to 15 at\%Al and Cu–Ni alloys up to 25 wt\%Ni. The Hall-Petch relation was found to be valid for all these alloys at 77, 194 and 293°K. The Petch slope \( k_y \) is nearly independent of temperatures in both alloy systems. Meanwhile, the concentration dependence of \( k_y \) differs considerably between these two alloy systems. The change in the Petch slope is nearly proportional to the square root of the concentration in Cu–Al alloys, while it seems to be proportional to the concentration itself in Cu–Ni alloys. The value of \( k_y \) depends strongly upon the solute concentration in Cu–Al alloys, but it does not in Cu–Ni alloys.

The results on Petch slope are not able to be explained by the break-away mechanism of a dislocation from the solute atmosphere with the help of the stress concentration by the pile-up dislocations. The examination of Li's model without the pile-up dislocations leads also to an unreasonable conclusion that the ledge density in the grain boundary exceeds the limiting value restricted by the lattice spacing. The results is also difficult to be explained by the conventional treatment of the creation mechanism of a dislocation. The creation mechanism is, however, essentially the same as the passage mechanism, in which the leading dislocation of the pile-up passes through the grain boundary leaving a misfit there. This mechanism seems to be able to explain the results of the present experiment. The quantitative comparison between the experiment and the calculation will be given in the following paper.

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