JOURNAL OF MATERIALS SCIENCE 18 (1983) 1881–1886

Temperature dependence of strength in a Cu_3Au–5% Ni alloy and its relevance to the APB morphology

AKITSUGU FUJITA*, YOSHINAO MISHIMA, TOMOO SUZUKI
Research Laboratory of Precision Machinery and Electronics, Tokyo Institute of Technology, Midori-ku, Yokohama 227, Japan

Temperature dependence of yield strength in a Cu_3Au–5%Ni alloy is investigated by compression tests over a temperature range between 77 and 700 K. The ordered alloy having $L1_2$ crystal structure exhibits swirl-like antiphase boundary morphology and was found not to possess the anomalous positive temperature dependence of strength. Nickel free Cu_3Au alloy has been reported to have the mechanical anomaly like Ni_3Al and to exhibit maze-like APB morphology. Such relevance between APB morphology and the temperature dependence of strength in $L1_2$ ordered alloys is discussed in terms of the relative phase stability between $L1_2$ and long period superlattice structures.

1. Introduction

The positive temperature dependence of yield strength has been observed in a number of $L1_2$ ordered alloys, for example in Ni_3Al [1, 2], Ni_3Ga [3], and Co_3Ti [4]. The thermally activated cross slip of screw dislocations onto cube planes, proposed by Kear and Wilsdorf [5], is the generally accepted mechanism for this anomalous mechanical behaviour in these alloys. In order for this mechanism to operate, antiphase boundary (APB) energy on (100) planes should be lower than that on (111) slip planes in the crystal structure. Flinn [6] had deduced that APB energy in $L1_2$ ordered alloys is lowest on the (100) plane when only the first nearest interaction is taken into account, and thereby, the increasing probability of occurrence of such a cross slip with increasing temperature can be expected in all of the alloys with the crystal structure. However, there are many $L1_2$ ordered alloys which exhibit normal temperature dependence of strength and, therefore, the anisotropy in APB energy must arise from more complicated reasons.

It has been recognized that thermally produced antiphase domain boundaries in $L1_2$ ordered alloys, having order–disorder transformation, can assume two types of morphologies, maze-like and swirl-like, as observed in thin foil transmission electron microscopy. The maze-like APB, square or rectangular networks in appearance, is observed in Cu_3Au [7–13], Au_3Cu [14, 15] and Cu_3Pt [16]. Trace analysis have shown that such networks consisted of (100) APBs indicating that the APB energy is lowest on (100) planes in these alloys. The swirl-like APB has been observed in Ni_3Fe [17–21] and Ni_3Mn [18, 22–24], where the morphology results from interfacial tension of APB domains and not from anisotropy in APB energies. Wee *et al.* [25] have shown that a Ni_3Fe single crystal shows no positive temperature dependence of flow stress associated with thermally activated cross slip mechanism.

Yodogawa *et al.* [16] have shown that the morphology of APB in Cu_3Au changes from maze to swirl-like when 5 at% Ni addition was made to the alloy. This was attributed by them to the reduction in the electron–atom ratio (e/a) of the alloy by the nickel addition and hence, to the increase in relative phase stability of the $L1_2$ structure against long period superlattice structures in which periodic (100) APB is characteristic. As was evidenced by Pope [26] and Kuramoto and

*Present address: Mitsubishi Heavy Industries, Nagaski 850-91, Japan.

0022–2461/83 $03.00 + .12

Pope [27], Cu_3Au does possess the positive temperature dependence of strength and, therefore, it is of interest to examine the relevance between the morphology of APB and the temperature dependence of strength in Cu_3Au–5%Ni as compared to that of Cu_3Au.

2. Experimental procedure

The alloy was prepared by arc melting using 99.99% alloying elements under an argon atmosphere. All ingot buttons were remelted together in a magnesia crucible with an electric furnace under the same atmosphere to obtain a uniform composition in a 27 mm diameter rod, which was then cold rolled to form 6 mm square rods.

Compression tests were carried out at temperatures between 77 and 700 K. Specimens, 5 mm in diameter and 10 mm in length, were cut from 5 mm diameter rods which were machined from a 6 mm square rod. An Instron-type testing machine was used for the test with a crosshead speed of $0.1\,mm\,min^{-1}$, giving a strain rate of approximately $1.67 \times 10^{-4}\,sec^{-1}$. Low temperature tests down to 77 K were performed by immersing the compression jig in an appropriate bath at a constant temperature, and elevated temperature tests up to 700 K were conducted in an argon atmosphere using a resistance furnace attached to the testing machine.

Compressive yield strength, as measured by 0.2% offset strength, was determined at various temperatures for both originally ordered and disordered alloys. Ordering heat-treatment prior to the test consists of; (a) annealing at 1000 K for 30 min followed by furnace cooling to 643 K, 20 K below order–disorder transformation temperature (T_c) of the alloy determined by Yodogawa *et al.* [16], (b) isothermal holding at this temperature for 85 h, and then (c) continuous cooling to 473 K at a rate of $2.5\,K\,hr^{-1}$ followed by air cooling to ambient temperature. Specimens of disordered state were obtained by annealing at 1000 K for 30 min followed by furnace cooling to 725 K, 60 K above T_c, and by water quenching after 10 min holding at this temperature. Specimens were placed in vacuum sealed silica tubes during each heat-treatment.

The morphology of APB in the ordered alloys was observed by a JEOL 200CX electron microscope operated at 200 kV. Thin foils were prepared by a twin-jet electropolisher using a CrO_3–CH_3COOH solution.

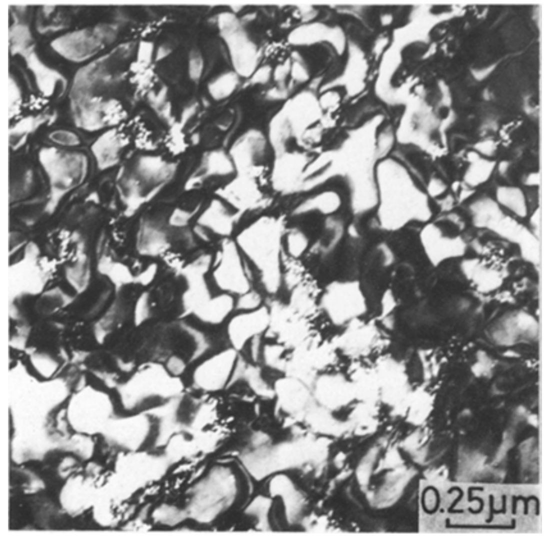

Figure 1 Thin foil electron micrograph of antiphase boundaries in ordered Cu_3Au–5%Ni alloy, showing a typical swirl-like APB morphology. Foil normal is ⟨1 1 0⟩.

3. Results and discussion

The morphology of APB in an ordered Cu_3Au–5%Ni alloy is shown in Fig. 1 exhibiting a typical swirl-like pattern, which is consistent with the results by Yodogawa *et al.* [16]. In their investigation they showed that the addition of nickel by up to 7% to a stoichiometric Cu_3Au alloy did not affect its single-phase $L1_2$ structure. This can be also predicted by the Au–Cu–Ni phase diagram determined by Raub and Engel [28]. It is ascertained that by an addition of 5%Ni to a Cu_3Au the morphology of APB is altered from maze to swirl-like without changing its fundamental crystal structure.

Results on compression tests for both the ordered and disordered Cu_3Au–5%Ni are given in Figs. 2 and 3. The temperature dependence of yield or flow stress in Cu_3Au alloys reported by Pope [26, 29], Langdon and Dorn [30], and Mohamed [31] are also shown in both figures. In comparing the present results on the Cu_3Au–5%Ni alloy to those previously reported for the Cu_3Au alloys, considerable solid solution hardening by a 5%Ni addition can be recognized. Fig. 2 shows the temperature dependence of strength in the initially disordered Cu_3Au–5%Ni alloy as well as those reported for the Cu_3Au alloys. The yield strength of the Cu_3Au–5%Ni alloy increases with increasing temperature at above 400 K in a manner closely resembling the data, by Mohamed [31], for a Cu_3Au alloy. Such hardening has

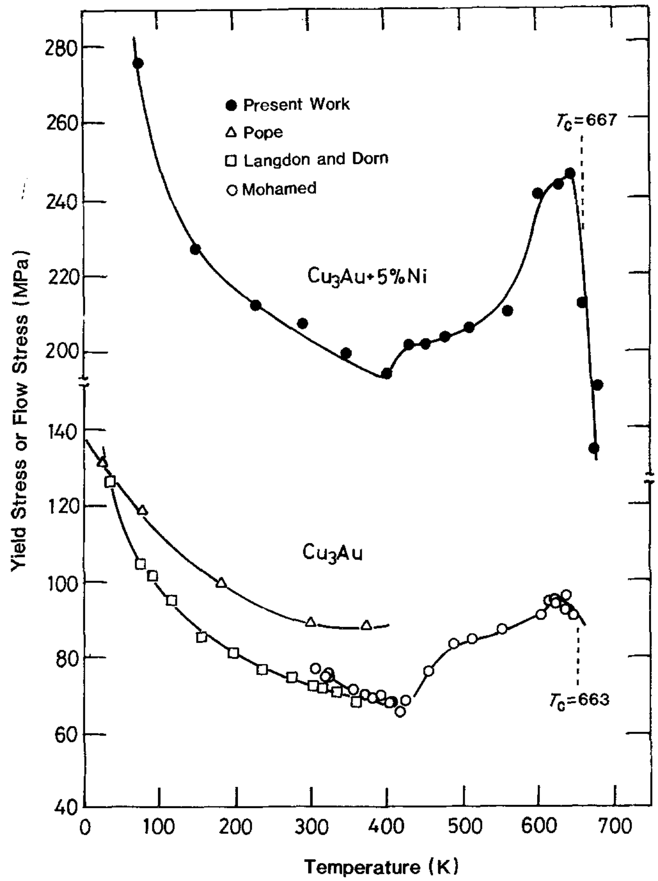

Figure 2 Temperature dependence of strength in a disordered Cu_3Au–5%Ni alloy. Data previously reported for Cu_3Au alloys by Pope [26], Langdon and Dorn [30] and Mohamed [31] are also shown.

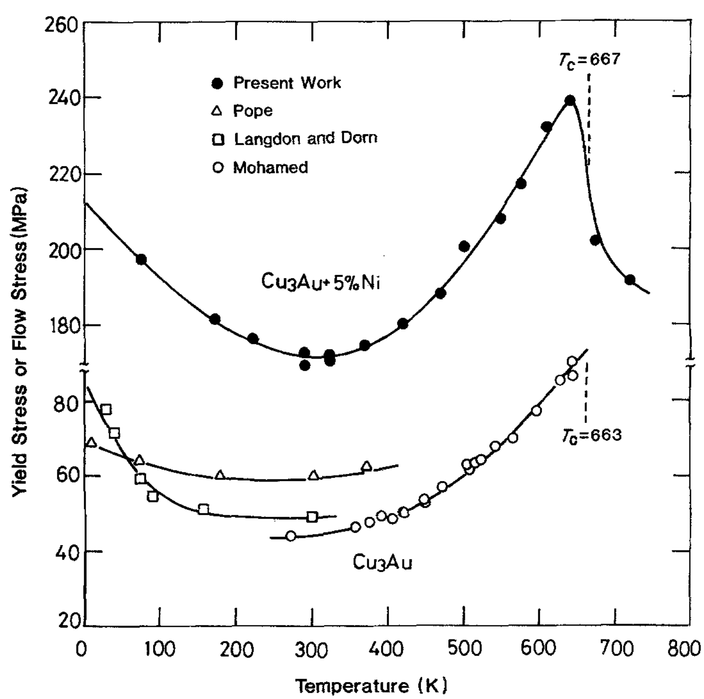

Figure 3 Temperature dependence of strength in an ordered Cu_3Au–5%Ni alloy. Data previously reported for Cu_3Au alloys by Pope [26], Langdon and Dorn [30] and Mohamed [31] are also shown.

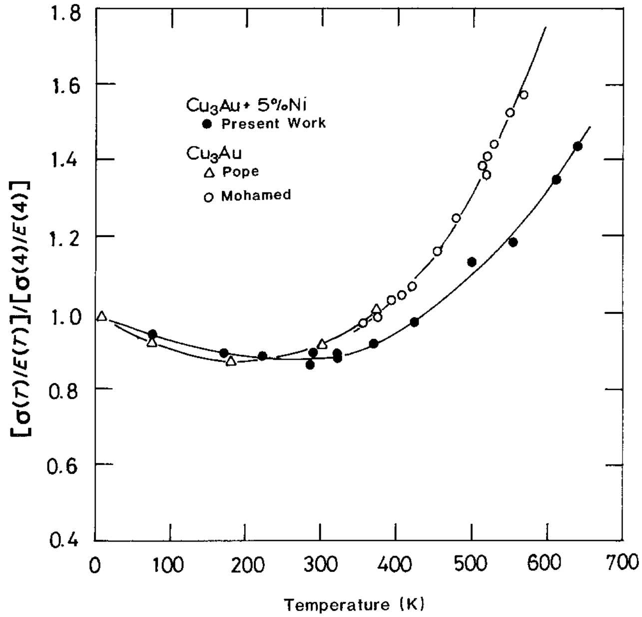

Figure 4 Temperature dependence of $[\sigma(T)/E(T)]/[\sigma(4)/E(4)]$ in an ordered Cu_3Au–5%Ni and Cu_3Au alloys [26, 31].

often been observed in an alloy having an order–disorder transformation temperature (T_c) below its melting point. The mechanism of hardening has been interpreted as the modulus interaction by Pope [26] or as ordering in the stress field of a dislocation by Sumino [32], which in either case is attributed to the state of mixture of the ordered and disordered state achieved by a diffusional process initially of ordering and then disordering as the temperature approaches T_c.

In Fig. 3, for initially ordered alloys of the $L1_2$ structure, the yield strength of the Cu_3Au–5%Ni alloy increases with increasing temperature from approximately 350 K until T_c is nearly reached. The temperature dependence of the yield strength or flow stress in Cu_3Au alloys is apparently very similar to that of the Cu_3Au–5%Ni alloy. In order to further discuss the temperature dependence of strength in both alloys, the effect of the temperature dependence of the elastic moduli has to be taken into consideration. A general expression on the critical resolved shear stress derived from the movement of a jogged dislocation in a single crystal can be given as;

$$\tau = \alpha \frac{G\mathbf{b}}{l}$$

where, G is the shear modulus, $\mathbf{b}$ is Burger's vector, l is the spacing between jogs and α is a constant. In the above expression, both τ and G are functions of temperature. Thus $\tau(T)/G(T)$ is the correct measure for the temperature dependence of the strength in a single crystal, and in the case for a polycrystal this parameter should be in the form of $\sigma(T)/E(T)$, where $\sigma(T)$ is the yield strength and $E(T)$ is Young's modulus measured at temperature T. In order to make a comparison of the present results on polycrystalline Cu_3Au–5%Ni alloy with that of the nickel free alloy, the yield strength data by Pope [26, 29] and Mohamed [31] are taken because their results can be represented together in a single curve only if each of Pope's data is lowered by 15 MPa. The data by Langdon and Dorn [30] are excluded because Pope [29] pointed out that the high impurity content in the material they used was responsible for the very strong temperature dependence of the strength at low temperatures. The $\sigma(T)/E(T)$ values are calculated for both alloys over the temperature range using the data on the variation of elastic modulus with temperature for Cu_3Au by Flinn *et al.* [33]. Each $\sigma(T)/E(T)$ value is then normalized by the value at 4 K, i.e. $\sigma(4)/E(4)$, which is obtained by extrapolating low temperature data. The temperature dependence of the yield strength of Cu_3Au–5%Ni and Cu_3Au alloys are plotted in terms of $[\sigma(T)/E(T)]/[\sigma(4)/E(4)]$ against temperature and are shown in Fig. 4. It becomes clear that the temperature dependence of strength in Cu_3Au–5%Ni and Cu_3Au is different in two ways. One is that the temperature at which the minimum strength is obtained is 350 K in the Cu_3Au–5%Ni alloy, whereas it is 250 K in the Cu_3Au alloy. The other is that the increase in strength with increasing temperature is more intense in Cu_3Au than in

Cu_3Au–5%Ni. Pope [26] has shown that the flow stress of ordered Cu_3Au was reversible at test temperatures up to 370 K, indicating no structural change of the ordered to disordered state below this temperature. It is also seen, in Fig. 2, that the increase in strength with increasing temperature in initially disordered alloy, which is attributed to the increasing portion of first ordered and then the disordered state through diffusional processes, occurs at above 400 K. Kuramoto and Pope [27] found that, in a single crystalline Cu_3Au alloy, the relative magnitude of the Schmidt factor for (1 0 0) to that of (1 1 1), known as the N value, increased with increasing temperature. This phenomenon had been observed in Ni_3Al, a typical $L1_2$ ordered alloy that exhibits a positive temperature dependence of strength. It can be concluded that the increase in strength with increasing temperature in Cu_3Au–5%Ni is attributed only to the state of mixture of the ordered and disordered regions achieved at above 350 K, the thermally activated cross slip mechanism is providing an additional increment in strength from low temperatures as in the case of Ni_3Al.

The present results would prove the relevance between APB morphology of an ordered $L1_2$ alloy and its temperature dependence of strength. Cu_3Au, that exhibits maze-like APB, does involve a thermally activated cross slip mechanism to provide the positive temperature dependence of strength, whereas Cu_3Au–5%Ni with swirl-like APB showed an increase in strength with increasing temperature only as a result of the order to disorder transformation giving rise to a state of mixture of the two states. The APB morphology change from maze to swirl-like in Cu_3Au by an addition of 5%Ni is attributed to the increase in the relative phase stability of the $L1_2$ structure against the long period superlattice structure by Yodogawa *et al.* [16]. The role of nickel in Cu_3Au is to decrease the electron–atom ratio of the alloy and, therefore, it can be speculated that the addition of an alloying element to increase the e/a of Cu_3Au, such as B-subgroup elements, would enhance the positive temperature dependence of the strength. When the relative stability of a long period superlattice structure increases by the increasing e/a value, (1 0 0) APB energy is expected to decrease so that thermally activated cross slip of a screw dislocation from (1 1 1) slip plane to (1 0 0) plane would become easier [34].

4. Conclusions

After investigating the temperature dependence of strength in a Cu_3Au–5%Ni alloy and its relevance to the APB morphology, the following conclusions can be drawn.

1. Compressive yield strength of a $L1_2$ ordered Cu_3Au–5%Ni alloy increases with increasing temperature at above 350 K till T_c is nearly reached.

2. This hardening is not related to the anomalous positive temperature dependence of strength which is associated with the thermally activated cross slip mechanism but to the state of mixture of ordered and disordered states achieved by a diffusional process as the temperature approaches to T_c.

3. The ordered Cu_3Au–5%Ni alloy exhibits a typical swirl-like APB morphology indicating that there is no anistropy in APB energy, in which the result is consistent with Yodogawa *et al.* [16].

4. A nickel free Cu_3Au alloy, known to exhibit maze-like APB, has been reported to possess the anomalous positive temperature dependence of strength. The change in morphology of the APB from maze to swirl-like by an addition of 5%Ni must have resulted in a loss of anistropy in the APB energy, and consequently in a loss of positive temperature dependence of the strength associated with the thermally activated cross-slip of a screw dislocation from (1 1 1) slip planes to (1 0 0) planes.

Acknowledgement

The present work is supported in part by the Grant-in-Aid for Scientific Research from the Ministry of Education, Science and Culture under Contract No. 57420051.

References

1. J. A. LOPEZ and G. F. HANCOCK, *Phys. Status Solidi (a)* **2** (1970) 469.
2. K. AOKI and O. IZUMI, *J. Jpn. Inst. Met.* **39** (1975) 1282.
3. S. TAKEUCHI and E. KURAMOTO, *J. Phys. Soc. Jpn.* **31** (1971) 1282.
4. P. H. THORNTON and R. G. DAVIES, *Met. Trans.* **1** (1970) 549.
5. B. H. KEAR and H. G. F. WILSDORF, *Trans. Met. Soc. AIME* **224** (1962) 382.
6. P. A. FLINN, *ibid.* **218** (1960) 145.
7. R. M. FISHER and M. J. MARCINKOWSKI, *J. Appl. Phys.* **31** (1960) 1687.
8. *Idem, Phil. Mag.* **6** (1961) 1385.
9. S. YAMAGUCHI, D. WATANABE and S. OGAWA, *J. Phys. Soc. Jpn.* **17** (1961) 1030.
10. M. J. MARCINKOWSKI and L. ZWILL, *Acta Metall.* **11** (1963) 373.

11. P. COULOMB and F. REYNAUD, *Mem. Sci. Rev. Met.* **118** (1971) 193.
12. M. SAKAI and D. E. MIKKOLA, *Met. Trans.* **2** (1971) 1635.
13. D. G. MORRIS, F. M. C. BESAG and R. E. SMALLMAN, *Phil. Mag.* **29** (1974) 43.
14. R. SINCLAIR, K. SCHNEIDER and G. THOMAS, *Acta Metall.* **23** (1975) 873.
15. D. GRATIAS and M. CONDAT, *C.R. Acad. Sci. Paris* **273** (1971) 336.
16. M. YODOGAWA, D. M. WEE, Y. OYA and T. SUZUKI, *Scripta Metall.* **14** (1980) 849.
17. P. COULOMB and F. REYNAUD, *C.R. Acad. Sci. Paris* **268** (1969) 912.
18. *Idem, Mem. Sci. Rev. Met.* **118** (1971) 193.
19. F. TOURAY, Y. CALVAYRAC and M. FAYARD, *C.R. Acad. Sci.* **272C** (1971) 1196.
20. Y. CALVAYRAC and M. FAYARD, *Mater. Res. Bull.* **7** (1972) 891.
21. D. G. MORRIS, G. T. BROWN, R. C. PILLER and R. E. SMALLMAN, *Acta Metall.* **24** (1976) 21.
22. P. COULOMB and F. REYNAUD, *C.R. Acad. Sci.* **266** (1968) 1107.
23. C. BOUYSSET, A. LASSERRE and P. COULOMB, *ibid.* **272C** (1971) 443.
24. F. REYNAUD, A. LASSERRE and P. COULOMB, *Acta Metall.* **23** (1975) 9.
25. D. M. WEE, O. NOGUCHI, Y. OYA and T. SUZUKI, *Trans. Jpn. Inst. Met.* **21** (1980) 237.
26. D. P. POPE, *Phil. Mag.* **25** (1972) 917.
27. E. KURAMOTO and D. P. POPE, *ibid.* **33** (1976) 675.
28. E. RAUB and A. ENGEL, *Z. Metallk.* **38** (1947) 11 and 147.
29. D. P. POPE, *Phil. Mag.* **27** (1973) 541.
30. T. G. LANGDON and J. E. DORN, *ibid.* **17** (1968) 999.
31. F. A. MOHAMED, *Acta Metall.* **26** (1978) 871.
32. K. SUMINO, *Sci. Rep. RITU* **A10** (1958) 283.
33. P. A. FLINN, G. M. McMANUS and J. A. RAYNE, *J. Phys. Chem. Solids* **15** (1960) 189.
34. H. SATO and R. S. TOTH, "Alloying Behavior and Effects in Concentrated Solid Solutions", edited by T. B. Massalski (Gordon and Breach Science Pub., New York, 1965).

Received 1 October
and accepted 23 November 1982