Ductile-to-Brittle Transition Characteristics in W–Cu Composites with Increase of Cu Content

Yutaka Hiraoka^{1,*1}, Takeshi Inoue^{1,*2}, Hideaki Hanado^{1,*2} and Naoyoshi Akiyoshi²

¹Department of Applied Physics, Okayama University of Science, Okayama 700-0005, Japan

²R&D Division, Toho Kinzoku Co., Ltd., Neyagawa 572-0036, Japan

A series of tungsten-copper composites containing copper of 19–80 vol% were subjected to three-point bend tests at temperatures between 77 and 363 K. Temperature dependences of the yield strengths and the maximum strengths were examined for each W–Cu composite. Then the effect of Cu content on the ductile-to-brittle transition behavior of the composite was investigated. The results are summarized as follows. First the composites containing copper of less than 40 vol% demonstrated ductile-to-brittle transition behavior and the transition temperature tended to decrease with the increase of Cu content. The composites containing copper of more than 60 vol%, in contrast, demonstrated no distinctive transition. Secondly production method leading to the microstructure had a significant effect on the maximum strength and consequently on the ductility, though it had almost no effect on the yield strength.

(Received February 4, 2005; Accepted May 2, 2005; Published July 15, 2005)

Keywords: tungsten-copper composite, production method, microstructure, mechanical properties, ductile-to-brittle transition

1. Introduction

Tungsten (W) is one of the transition metals with a body-centered cubic (BCC) lattice. Tungsten has the highest melting point among metals. In common the BCC metals demonstrate a transition phenomenon from ductile to brittle with the decrease of test temperature. The temperature showing such a transition is called “ductile-to-brittle transition temperature (DBTT)”. DBTT of tungsten after the complete recrystallization is much higher than room temperature. Hence the material fractures completely in a brittle manner.¹⁾ On the other hand, copper (Cu) is one of the metals with a face-centered cubic (FCC) lattice. In contrast to the BCC metals, the FCC metals generally demonstrate no distinctive ductile-to-brittle transition phenomenon and excellent ductility at room temperature. In addition copper has a very low melting point.

Tungsten-copper composites (W–Cu composites) are widely used as the electric and electronic materials such as heavy-duty electrical contacts, high-current circuit breakers, resistance welding electrodes and contact tips of arc welding guns.^{2–5)} Until now many researchers investigated the mechanical properties of the W–Cu composites. It is summarized that both the hardness and the strength of the composite depend on both the microstructure and the chemical composition.^{2,4,6–12)} First the hardness approximately obeys the rule of mixtures. The hardness increases almost linearly with increasing the volume fraction of tungsten phase in the matrix of copper. Secondly the production method significantly influences the mechanical properties of the composite. Generally the composite produced by infiltrating the tungsten powder skeleton with liquid copper demonstrates much superior properties than that by pressing and sintering the mixed powders of tungsten and copper. Thirdly the fracture toughness of the composite is improved with the increase of copper content through the

preferential deformation of the copper phase. However report concerning the ductile-to-brittle transition characteristics in the W–Cu composite with the increase of copper content is only limited. In this work we performed the three-point bend tests at temperatures between 77 and 363 K for the W–Cu composites containing copper of 19–80 vol%. These composites were produced alternatively by infiltrating the tungsten powder skeleton with liquid copper or by pressing and sintering the mixed powders of tungsten and copper. First the changes of strength and ductility of the composite as a function of the test temperature and the copper content were investigated. Secondly the effects of copper content on the ductile-to-brittle transition behavior of the composite were discussed from the viewpoints of the microstructure and the fractography.

2. Experimental Procedures

2.1 Materials and specimen preparation

The composites were produced by two kinds of methods. One is by infiltrating the tungsten powder skeleton with liquid copper. The other is by pressing and sintering the mixed powders of tungsten and copper. The composite containing copper of 19, 27, 35 and 48 vol% was produced by the former method and is designated as W19Cu(a), W27Cu(a), W35Cu(a) and W48Cu(a), respectively. On the other hand, the composite containing copper of 20, 40, 60 and 80 vol% was produced by the latter method and is designated as W20Cu(b), W40Cu(b), W60Cu(b) and W80Cu(b), respectively. The porosities in the composites were as low as 2–4% except for W80Cu(b) (about 7%). Typical starting dimension of the composite was 25 × 50 × 4 mm. Commercial pure copper (designated as “OFHC-Cu”) sheet of 1 mm thick and commercial pure tungsten sheet of 1 mm thick were used for the reference materials.

2.2 Microstructure observations

Rectangular specimens were cut put from the composite materials. The surfaces of the specimen were polished using

*1Corresponding author, E-mail address: hiraoka@dap.ous.ac.jp

*2Graduate student, Okayama University of Science

Fig. 1 Typical microstructure of W-Cu composites.

emery papers and buff clothes. The polished surfaces of the composites were observed using SEM. Typical microstructures are shown in Fig. 1. The bright and gloomy images in the photographs are W-phase and Cu-phase, respectively. The microstructures of W19Cu(a) and W20Cu(b) are similar.

The Cu-phases are apparently isolated in the matrix of tungsten. Such a microstructure suggests that the W-W contiguity is primary. With the increase of Cu content, interconnection of the Cu-phases becomes much more frequent and hence the W-W contiguity decreases. Finally,

in W60Cu(b) and W80Cu(b), W-phases are generally isolated in the matrix of copper. The microstructure of W48Cu(a) is just intermediate between that of W35Cu(a) (W40Cu(b)) and that of W60Cu(b).

2.3 Three-point bend tests

From the composite material, bend-test specimens of 4 mm wide, 25 mm long and 1.5–2.0 mm thick were cut out. The surfaces of the specimen were polished using emery papers. Three-point bend tests were carried out at the temperature range between 77 and about 363 K at a crosshead speed of 0.011 mm/s. From the load-displacement curve at each temperature, the yield (σ_y) and maximum (σ_m) strengths and the plastic strain (ε) were calculated using the following equations.

$$\sigma_y(\sigma_m) = \frac{3aP_y(P_m)}{wt^2} \quad (1)$$

$$\varepsilon = \frac{6tx}{(2a)^2} \quad (2)$$

w (mm) and t (mm) are the specimen width and thickness, respectively. $2a$ (= 16 mm) is the span of the supporting pins. P_y and P_m are the yield point load and the maximum load, respectively. x (mm) is the displacement of the crosshead prior to failure. In this work the maximum displacement was mechanically limited ($x_{\max} = 6$ mm).

2.4 Fracture surface observations

The fracture surface of the composite after the test was observed using SEM. The general fracture mode and the manner of crack generation and propagation were examined. The grain boundary of the W-phase and the interface between W-phase and Cu-phase were identified.

2.5 Definition of critical stress and critical temperature

From the change of the yield and maximum strengths as a function of test temperature, two parameters, critical stress (σ_c) and critical temperature (T_c) can be obtained.^{13–15} Definition of these parameters is schematically shown in Fig. 2. The critical stress is the stress that propagates micro-crack(s) alternatively along the grain boundaries and/or in the matrix. This stress represents the experimental fracture strength at low temperatures. The critical temperature, on the other hand, is so-called DBTT or the nil-ductility transition temperature (NDT). The reciprocal of the critical temperature is a measure of low-temperature ductility.

3. Results

3.1 Bend strength and ductility

The changes of the yield and maximum strengths as a function of test temperature are shown in Fig. 3 for the composite by the infiltrating method and in Fig. 4 for the composite by the pressing and sintering method. In general the yield strength is approximately a linear function of the reciprocal of temperature ($1/T$). The composite containing copper of less than 40 vol% demonstrated a ductile-to-brittle transition behavior. Namely the specimen failed in a brittle manner at a given low temperature, though the specimen demonstrated small amounts of plastic strain even at room

Fig. 2 Definition of two parameters, critical stress and critical temperature.

temperature. With the increase of Cu content, the slope of the linear relation between the yield strength and the reciprocal of test temperature tended to decrease. Consequently the yield and the maximum strength did not intersect each other. The composites demonstrated a certain amount of plastic strain even at a temperature as low as 77 K.

It is well known that some physical properties like electrical resistivity, thermal expansion ratio and hardness obey the rule of mixture. In Fig. 5 the yield strength at room temperature of the composite is plotted as a function of Cu content. Data of the pure tungsten and OFHC-Cu are also plotted in the figure. The pure tungsten specimen was subjected to the heat treatments alternatively at 1373 K (designated as “SR-tungsten”), at 1573 K (“R1-tungsten”) or at 1773 K (“R2-tungsten”). Pure tungsten specimen demonstrated no plastic strain at room temperature. Thereby the yield strength at room temperature in the figure was obtained by extrapolating the yield strengths at higher temperatures to that at room temperature.¹⁶⁾ It is shown that the data of the composite by the pressing and sintering method approximately obeyed the rule of mixture demonstrated by the straight line. However, the data of the composite by the infiltrating method slightly deviated downward from the straight line.

In Fig. 6 the maximum strength at room temperature of the composite is plotted as a function of Cu content. Data for the pure tungsten¹⁶⁾ and OFHC-Cu¹⁷⁾ are also plotted in the figure. The maximum strength of the composite containing copper of 19–48 vol% by the infiltrating method was as high as 1400–1500 MPa. This value is as high as the strength of SR-tungsten and much higher than that of R1- or R2-tungsten. In contrast the strength of the composite containing copper of 20–40 vol% by the pressing and sintering method was generally much lower than that of the composite by the infiltrating method. For the composite containing copper of 60–80 vol%, on the other hand, the maximum strength tended to decrease with the increase of Cu content.

In Fig. 7 the plastic strain at room temperature of the composite is plotted as a function of Cu content. Data for the

Fig. 3 Change of yield and maximum strengths as a function of test temperature for the composite produced by infiltrating method.

Fig. 4 Change of yield and maximum strengths as a function of test temperature for the composite produced by pressing and sintering method.

pure tungsten¹⁶⁾ are also plotted in the figure. The pure tungsten demonstrated no plastic strain at room temperature. However the composite demonstrated a certain extent of

plastic strain. For the composite containing copper of 19–35 vol% by the infiltrating method, the plastic strain was small but tended to increase with the Cu content. In this

Fig. 5 Plots of yield strength as a function of Cu content.

Fig. 8 Plots of critical stress as a function of Cu content.

Fig. 6 Plots of maximum strength as a function of Cu content.

Fig. 7 Plots of plastic strain as a function of Cu content.

compositional range the plastic strain of the composite by the pressing and sintering method was generally smaller than that of the composite by the infiltrating method. The plastic strain of the composite containing copper of 48–80 vol%, on the other hand, is generally much larger irrespective of the production method. It is noted that the composite demonstrating large plastic strain corresponds to that demonstrating no distinctive ductile-to-brittle transition in a manner similar

to OFHC-Cu. OFHC-Cu did not fail until the maximum displacement¹⁷⁾ and hence the data was not shown in the figure.

3.2 Critical stress and critical temperature

The critical stress of the composite is plotted as a function of Cu content in Fig. 8. The critical stresses of the pure tungsten¹⁶⁾ are also plotted in the figure. It is noted that the critical stress of the pure tungsten significantly depends on the microstructure such as grain size. The critical stress of the composite containing copper of 19–35 vol% by the infiltrating method is as high as 1500 MPa. This value is relatively larger than that of R1- or R2-tungsten and as high as that of SR-tungsten. In addition the critical stress of the composite by the pressing and sintering method was generally much lower than that of the composite by the infiltrating method.

The reciprocal of critical temperature of the composite is plotted as a function of Cu content in Fig. 9. The reciprocal of the critical temperature is a measure of low temperature ductility of the material as already mentioned in the section 2.5. The critical temperatures of the pure tungsten¹⁶⁾ are also plotted in the figure. It is noted that the effect of microstructure such as grain size on the critical temperature in the pure tungsten was not significant in contrast to the critical

Fig. 9 Plots of reciprocal of critical temperature as a function of Cu content.

Fig. 10 Typical fractograph of W-Cu composites by infiltrating method.

stress. Low temperature ductility (reciprocal of the critical temperature) of the composite by the infiltrating method increased substantially with the increase of Cu content. The ductility of the composite by the pressing and sintering method was generally much lower than that of the composite by the infiltrating method.

3.3 Fractography

Typical fractograph is shown in Fig. 10 for the composite by the infiltrating method and in Fig. 11 for the composite by the pressing and sintering method. The fracture surface of the composite containing copper of 19–40 vol% basically consisted of two kinds of interfaces. One is the grain boundary or grain boundaries of the W-phase, namely W-W contiguity (marked by “A” in Fig. 10(1)). The other is the interface(s) between W-phase and Cu-phase (marked by “B”). It is considered the latter interface(s) is not so strong, since tungsten and copper are insoluble each other and hence make no alloys. Besides these interfaces, well-deformed Cu-phase is also observable (marked by “C”). It is obvious that “A” and “B” interfaces are surrounded by “C”. It is noted, first, both the ratio and the size of W-W contiguity tends to decrease with the increase of Cu content. Secondly the ratio of the well-deformed Cu-phase is generally less in the composite by the pressing and sintering method than in the composite by the infiltrating method.

The fractography of the composite containing Cu of more than 48 vol% was generally different from that of the composite containing copper of less than 40 vol%. The W-W contiguity was rarely observable and the deformation of the Cu-phase was much more distinguished.

4. Discussion

4.1 Effect of Cu content on yield strength

Micro-hardness of the composite well obeyed the rule of mixture as expressed by the equation.¹⁷⁾

$$H_v = 400(1 - V_{Cu}) + 70V_{Cu} \quad (3)$$

H_v is the microhardness number and V_{Cu} is the volume fraction of Cu-phase in the composite. It can be expected that yield strength of the composite also obeys the rule of mixture. The data for the composite by the pressing and sintering method almost obeyed the rule of mixture. However the data of the composite by the infiltrating method deviated, though slightly, downward from the rule. The disagreement between the microhardness and the yield strength might be attributed to the microstructure of the composite. In the composite containing lower Cu content, dispersion of the Cu-phase in the matrix of tungsten seems to be mostly homogeneous. In the composite by the infiltrating method, however, the W-phase might be surrounded by the three-dimensional network of the Cu-phase, since the porosity is as low as 2–4%. In this case it is expected preferred and localized deformation of the Cu-phase occurs and consequently downward deviation from the rule of mixture appears.

4.2 Effect of Cu content on critical stress

As already mentioned in the section 2.5, critical stress is the stress which represents the experimental fracture strength at low temperatures. The critical stress of W19Cu(a), W27Cu(a) and W35Cu(a) by the infiltrating method was as high as that of the pure tungsten after stress-relieving

Fig. 11 Typical fractograph of W–Cu composites by pressing and sintering method.

(relatively fine grained) and much higher than that after recrystallization (relatively coarse grained). These results are interpreted with the characteristic microstructure of the composite that W-phase is always surrounded by the Cu-phase. In this case propagation of the crack(s) which generates from the W/Cu and/or W/W interfaces is effectively suppressed by the local deformation of the Cu-phase. Mu *et al.*¹¹⁾ reported that fracture toughness of the W–Cu composite is substantially improved with the increase Cu content through a preferential plastic deformation of the Cu-phase surrounding the W-phase.

On the other hand, the composite by the pressing and sintering method demonstrated much lower ductility than that by the infiltrating method. The parameters, critical stress and critical temperature were obtainable only for W40Cu(b). This result is interpreted as follows. First the strengths of the W/Cu and/or W/W interfaces are relatively low in the composite by the pressing and sintering method. In the composite by the infiltrating method, higher sintering temperature to prepare the tungsten powder skeleton should lead to stronger W/W interfaces. Secondly the W-phase in the composite by the infiltrating method is always surrounded by the infiltrated Cu-phase, even though the Cu-phase looks apparently isolated in the W-phase matrix. In contrast the W-phase in the composite by the pressing and sintering method may not be always surrounded by the Cu-phase. The latter microstructure suggests that propagation of the crack cannot be effectively suppressed by the Cu-phase and leads to lower fracture strength with large scattering.

4.3 Ductile-to-brittle transition characteristics

Some of the composites demonstrated a ductile-to-brittle transition behavior and hence critical temperature was obtainable as already shown in Fig. 9. The results are summarized as follows. First the ductility of the composite was generally much higher than that of the pure tungsten after recrystallization. Secondly the ductility of the composite tended to increase with the increase of Cu content. Thirdly the ductility of the composite by the infiltrating method was generally higher than that by the pressing and sintering method. These results are interpreted as follows. As mentioned already in the section 2.5, the critical temperature, namely the low temperature ductility, depends both on the maximum strength (critical stress) and the yield strength. Thereby the higher ductility of the composite than that of the pure tungsten after recrystallization is attributed not only to the higher maximum strength but to the lower yield strength as shown in Figs. 5 and 6. Further improvement of the ductility with the increase of Cu content is attributed mainly to the linear decrease of the yield strength with the increase of Cu content. Lastly lower ductility of the composite by the pressing and sintering method than that of the composite by the infiltrating method is attributed mostly to the lower maximum strength of the former composite.

Ductile-to-brittle transition behavior was also recognized as a function of Cu content. The composite containing copper of less than 40 vol% fractured in a brittle manner at low temperatures, though the composite demonstrated a certain extent of plastic deformation at higher temperatures. Such a transition is one of the characteristics for BCC metals like the

pure tungsten. On the other hand, the composite containing copper of more than 48 vol% did not demonstrate such a transition in a manner similar to the pure copper which is one of typical FCC metals. High ductility of the composite containing copper of more than 48 vol% is attributed not only to the lower yield strength but to the less temperature dependence of the yield strength. In the composite containing high Cu content, preferential deformation of the Cu-phase occurs with the W-phase being not deformed. Belk *et al.*¹⁰⁾ reported that even cold work of thickness reduction of 25% induces confined deformation of Cu-phase. Hiraoka *et al.*¹²⁾ also recently reported that morphology of W-particle in W-80 vol%Cu composite is not changed after plastic deformation of 10% at room temperature. From the present result it is concluded that the critical Cu content for the ductile-to-brittle transition behavior is 40–48 vol%.

5. Conclusions

- (1) W–Cu composite containing copper of 19–40 vol% demonstrated a ductile-to-brittle transition behavior in a manner similar to the pure tungsten. Critical stress increased and critical temperature decreased with the increase of Cu content.
- (2) Both the low temperature fracture strength and ductility of the composite by the pressing and sintering method were generally lower than those by the infiltrating method.
- (3) W–Cu composite containing copper of more than 48 vol% demonstrated no distinctive transition in a manner similar to the pure copper. This result is attributed not only to the lower yield strength but to the less temperature dependence of the strength.
- (4) Critical Cu content for the ductile-to-brittle transition is 40–48 vol%.

Acknowledgment

We greatly appreciate to Toho Kinzoku Co., Ltd. for supplying W–Cu composite.

REFERENCES

- 1) T. E. Tiez and J. W. Wilson: *Behavior and Properties of Refractory Metals* (Univ. Tokyo Press, 1965).
- 2) K. V. Sebastian: *Int. J. Powder Metall. & Powder Technol.* **17** (1981) 297–303.
- 3) M.-H. Hong, S. Lee, E.-P. Kim, H.-S. Song, J.-W. Noh and Y.-W. Kim: *Proc. of the 13th Int. Plansee Sem.* (Plansee AG, Reutte-in-Tirol, 1993) pp. 451–460.
- 4) K. Wojtasik and S. Stolarz: *Proc. of the 13th Int. Plansee Sem.* (Plansee AG, Reutte-in-Tirol, 1993) pp. 471–478.
- 5) W. S. Wang and K. S. Kwang: *Metall. Trans. A* **29A** (1998) 1509–1516.
- 6) I.-H. Moon, S.-S. Ryu and J.-C. Kim: *Proc. of the 14th Int. Plansee Sem.* (Plansee AG, Reutte-in-Tirol, 1997) pp. 16–26.
- 7) B. Bryskin and E. K. Ohriner: *Proc. of the 14th Int. Plansee Sem.* (Plansee AG, Reutte-in-Tirol, 1997) pp. 358–370.
- 8) A. A. Sadek, M. Ushio and F. Matsuda: *Metall. Trans. A* **21A** (1990) 3221–3235.
- 9) B. L. Mordike, J. Kaczmar, M. Kielbinski and K. U. Kainer: *Powder Metall. Int.* **23** (1991) 91–95.
- 10) J. A. Belk, M. R. Edwards, W. J. Farrell and B. K. Mullah: *Powder Metall.* **36** (1993) 293–296.
- 11) K. Mu, Y. Kuang, G. Xu and A. Wei: *Proc. of the 14th Int. Plansee Sem.* (Plansee AG, Reutte-in-Tirol, 1997) pp. 381–392.
- 12) Y. Hiraoka, H. Hanado and T. Inoue: *Int. J. Refr. Met. & Hard Mater.* **22** (2004) 87–93.
- 13) A. S. Wronski, A. C. Chilton and E. M. Capron: *Acta Metall.* **7** (1969) 751–755.
- 14) Y. Hiraoka, S. Yoshimura and K. Takebe: *Int. J. Refr. Met. & Hard Mater.* **12** (1993–1994) 261–268.
- 15) S. Yoshimura, Y. Hiraoka and K. Takebe: *J. Japan Inst. Met.* **58** (1994) 734–739 (in Japanese).
- 16) Y. Hiraoka and H. Kurishita: unpublished work.
- 17) Y. Hiraoka: unpublished work.