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Microstructural characteristics in tough pitch copper for revealing the work hardening region

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Abstract. To reveal localized plastic deformation zones in a tough pitch copper, the etching characteristics of a copper sample have been examined. The etching was carried out on a sample surface using an etchant consisting of 25 ml nitric acid solution and 75 ml water. To clarify the plastic deformation zone, the sample deformed plastically was heated to between 250°C and 300°C before the etching process. This is due to a change of the microstructure and crystal orientation in the plastic deformation zone producing recrystallized small grains. In this case, the plastically deformed zone is severely etched, whereas the undeformed zone is only slightly etched. Identification of the details of the deformation zone from the etching is further discussed.

Keywords: etching technique; tough pitch copper; plastic deformation; microstructure

1. Introduction

In recent years, tough pitch copper has been employed for various engineering applications, such as wires, tubes and several other products. The use of tough pitch copper for engineering applications requires an examination of its mechanical properties. Prasad and Rao have investigated the stress vs. strain relations of tough pitch copper, which exhibit flow softening with single or multiple peaks before the flow stress reaches a steady state (Prasad *et al.* 2006). To make high strength tough pitch copper, the cold working process has been studied over a wide range of strain rate. With torsion under high pressure and drawing (an equal channel angular pressing technique), ultrafine-grained copper in bulk is created, which leads to high tensile and high fatigue strength. In the study by Kunz *et al*. (Kunz *et al*. 2006), the fatigue lifetime of ultrafine-grained Cu of 99.9% is shown to exceed that of its conventional grained cold worked counterparts by a factor of 1.7. Although much information regarding the fatigue properties of tough pitch copper has been reported, there is apparently a lack of understanding of the details of how to induce failure characteristics during cyclic loading. In general, the fatigue strength is attributed directly to the

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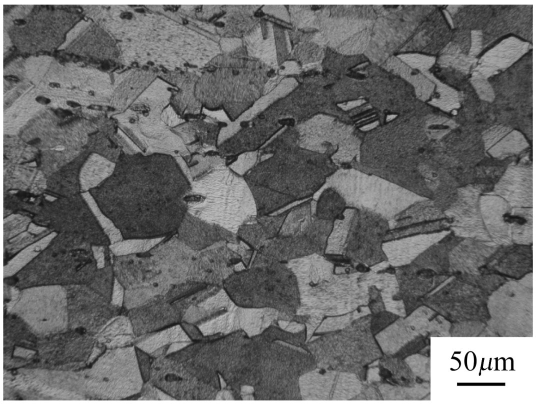

Fig. 1 Optical micrograph of tough pitch copper

crack growth rate, which is affected by the severity of plastic deformation around the crack tip.

The effect of plastic deformation on the crack growth characteristic has been studied using various experimental techniques, e.g. finite element analysis, moiré camera, photoelastic stress analysis, microhardness measurement and etching techniques. As the plastic deformation zone can be identified easily and accurately, the etching technique is a suitable approach for observing plastically deformed zones. Fry (Fry 1921) and Morris (Morris 1949) have proposed using etching techniques, in which the plastic deformation zones in mild steels can be revealed, because the segregation of impurity atoms occurs at dislocation cores, i.e., etch-pit formation. Recently, one of the authors has proposed using etching techniques to clarify the plastic deformation zones for some metals (Okayasu *et al*. 2005, Okayasu *et al*. 2008, 2011).

To date, there have been several etching techniques proposed for various engineering materials, such as mild steel, carbon steel, stainless steel, aluminum alloy and magnesium alloy (Fry 1921, Morris 1949, Okayasu *et al*. 2005, Okayasu *et al*. 2008, 2011). However, there is no clear etching process for copper. The purpose of this work is therefore to propose a new etching technique to detect the localized plastic deformation zone in tough pitch copper. Moreover, an investigation has been conducted to understand the detailed etching mechanism that identifies the deformation zone.

2. Experimental

2.1 Material and experimental procedures

In the present work, a tough pitch copper (Cu: 99.96%) was employed. Fig. 1 shows the microstructure of the tough pitch copper observed by an optical microscope. The nominal grain size measured is 89 μm. The tensile properties of the tough pitch copper at room temperature are: tensile strength σ_{UTS} = 195 MPa and strain to the failure ε_f = 35%. The specimens were machined into the form of a rectangular block (10 × 10 × 5 mm). A severe plastic deformation was introduced by imposing a hardened high carbon steel wire with diameter 0.8 mm at a compressive loading to approximately 3.0 kN. A screw driven type universal testing machine with 10 kN capacity was employed to conduct the compressive loading.

Table 1 Heating conditions for tough pitch copper

	Temp. - time		Temp. - time
Sample A	no heating	Sample G	250°C - 8 h
Sample B	180°C - 2 h	Sample H	300°C - 2 h
Sample C	180°C - 8 h	Sample I	300°C - 8 h
Sample D	220°C - 2 h	Sample J	400°C - 2 h
Sample E	220°C - 8 h	Sample K	500°C - 2 h
Sample F	250°C - 2 h	Sample L	600°C - 2 h
		Sample M	700°C - 2 h

In order to identify the plastic deformation zone by the etching technique, the plastically deformed samples were heated to various temperatures between 180°C and 700°C, as summarized in Table 1. The heating conditions were determined on the basis of the recrystallization temperature for the tough pitch copper, which is about 230°C (Sudo *et al.* 1997). The aim of the heating process is to reveal the different microstructural characteristics between the undeformed zone and plastic deformation zone. The etching technique used to identify the deformation zone is briefly summarized as follows: (1) the specimens are compressed to make a severe plastic deformation, (2) the deformed samples are heated to different temperatures with different heating times before air cooling, (3) the specimen surface to be observed is ground to a mirror finish and (4) the mirror face is etched for 300 sec using an etchant consisting of 25 ml 60%-nitric acid solution and 75 ml water. In this case, the following chemical reaction occurs: $8HNO_3 + 3Cu \rightarrow 3Cu(NO_3)_2 + 4H_2O + 2NO$.

2.2 Microstructural observation

Microstructural characteristics of the tough pitch copper were investigated by an optical microscope and electron backscatter diffraction (EBSD). The sample surfaces for all observation were polished to mirror flatness in a vibropolisher using colloidal silica. The electron backscatter diffraction analysis was executed using a high resolution electron JSM-7000F microscope (JEOL Ltd.) with the following conditions: beam current 5 nA, an accelerating voltage of 15 kV and step size 1 μm. The EBSD analysis of the microstructure is resolved into 42860 image pixels. Based upon the analysis of the EBSD patterns, the crystal orientations of the samples were analyzed using HKL Channel 5 software. Details of the EBSD technique are described in Ref. (Randle 2003).

2.3 Finite element analysis

Finite element analysis (FEA) was performed to analyze the plastic deformation pattern in the specimen material. In this analysis, two-dimensional finite element simulation with 8-noded quad elements was employed. A FEA model in the present study was designed based upon the specimen as described above. The mesh size of the specimen surrounding the wire was designed to be 0.25 mm. The ANSYS 12.1 program was used in this calculation, where the analysis was conducted to examine the von-Mises plastic strain distribution under plane stress criteria. The material properties of the specimen materials were: elastic constant $E = 128$ GPa, Poisson's ratio $\nu = 0.33$,

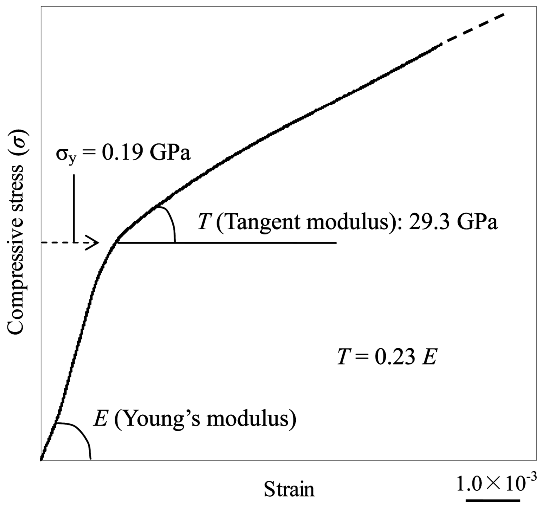

Fig. 2 Compressive stress vs. strain relation of tough pitch copper

tangent modulus T = 29.3 GPa and yield strength σ_y = 0.19 GPa. In this study, the tangent modulus was obtained from the compressive stress vs. compressive strain relation, examined experimentally (see Fig. 2), where compressive stress was conducted using a specimen (2.5 × 2.5 × 4.5 mm) at 1.0 mm/min. Because the hardness for the hardened wire is much higher than the tough pitch copper, e.g. about 600% high, the model of the wire was designed to be rigid.

3. Results and discussion

Fig. 3(a) displays the macrostructure of the etched samples in the area around the dented zone. It is clear that a round bright (white) region in the vicinity of the dented zone was similarly observed for the samples heated to 250°C-2 h, 250°C-8 h and 300°C-2 h (Samples F, G and H). The depth of the white area is approximately 1.0 mm for the three samples, as indicated in Fig. 3(a). On the other hand, the bright areas cannot be observed in the other samples. In this case, the bright round zones adjacent to the dented areas may be attributed to severe etching. To verify this, further examination of the chemical reaction to reveal the bright zone was conducted, where dented zone was further made on the other face of the sample for 300°C-2 h. Fig. 3(b) displays the obtained macrograph and that shown with a high magnification. As seen, clear bright round zones are detected in both top and bottom of the sample around the dented zones, which are similar to that in Fig. 3(a).

Fig. 4 presents an optical micrograph showing the microstructure of Sample H near to and far from the dented zone. Note that the white round area, shown in Fig. 3, corresponds to a dark zone (Area A) in Fig. 4. Due to the lighting conditions, the etched zones appear white and dark in the photographs taken by a digital camera (Fig. 3) and optical microscope (Fig. 4), respectively. As can be seen in Fig. 4, different microstructural characteristics are detected. For example, the grain size is smaller in the sample adjacent to the dented zone. It is also clear that with the etchant solution, the greatest corrosion occurs at the grain boundary, as schematically illustrated in Fig. 4. In previous reports, similar profiles of the round areas can be obtained around the dented region, which were

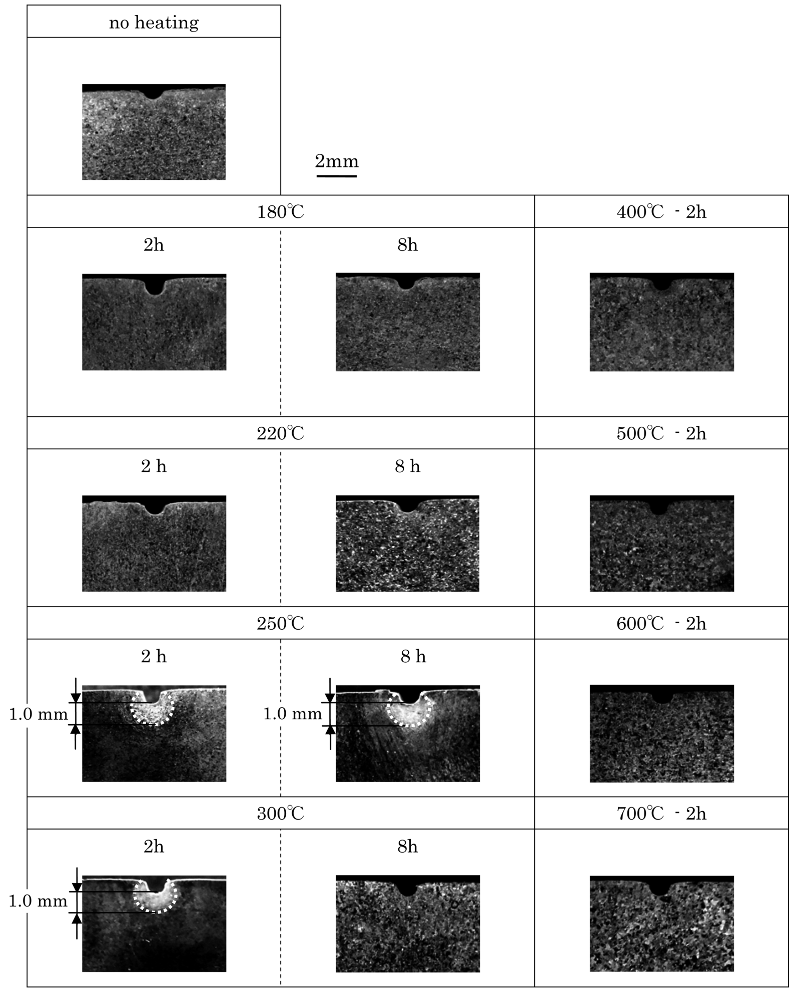

Fig. 3(a) Optical macrographs of the samples after the etching technique. The white region is the deformed zone shown by the dotted line

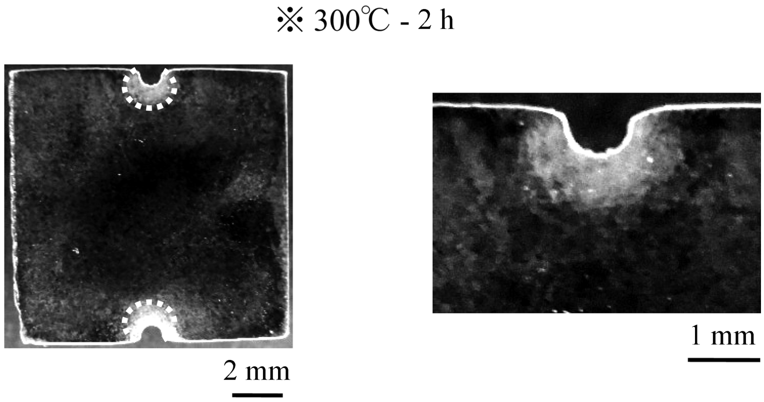

Fig. 3(b) Optical macrographs of the sample (300ºC - 2 h) after the etching technique

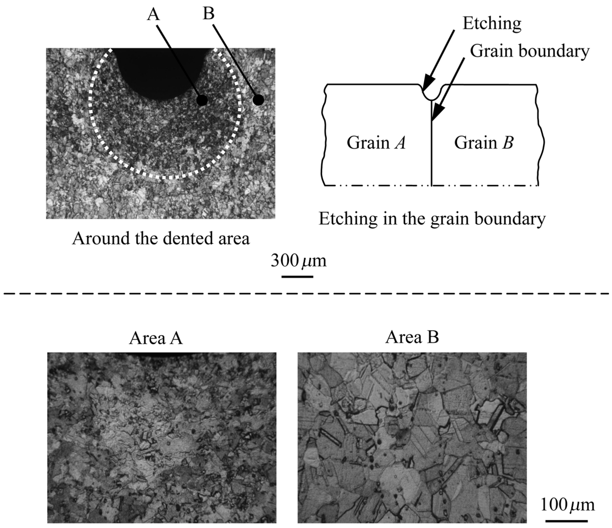

Fig. 4 Optical micrographs of undeformed and deformed regions in Sample H

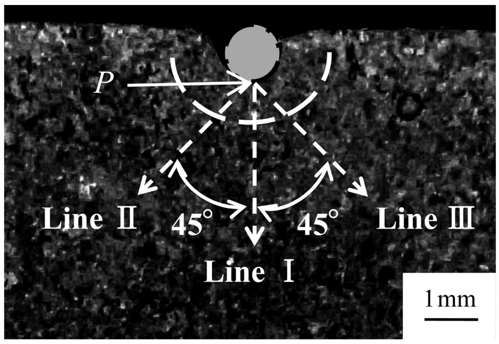

Fig. 5 Directions of microhardness measurement indicated by arrows

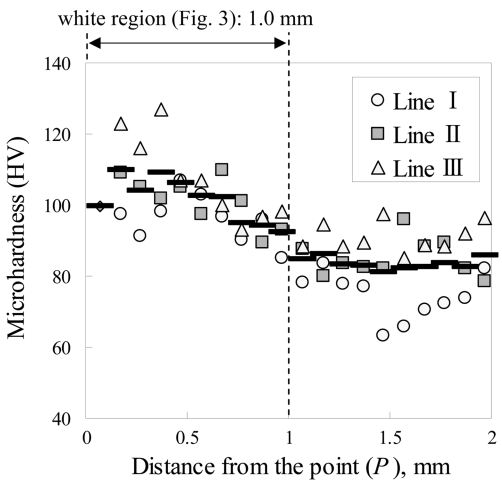

Fig. 6 Microhardness results from Sample A, measured from position *P*, as shown in Fig. 5

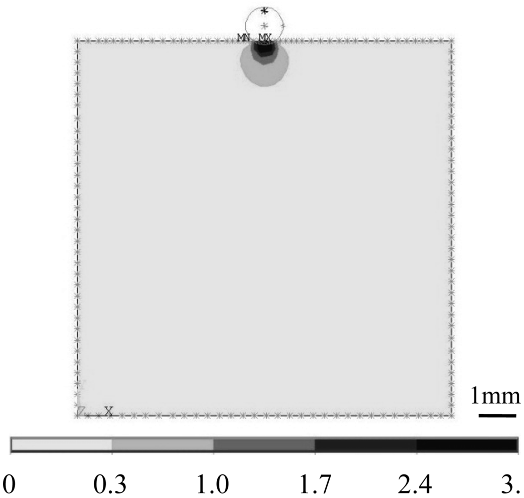

Fig. 7 von-Mises plastic strain distribution revealed via FE analysis for tough pitch copper

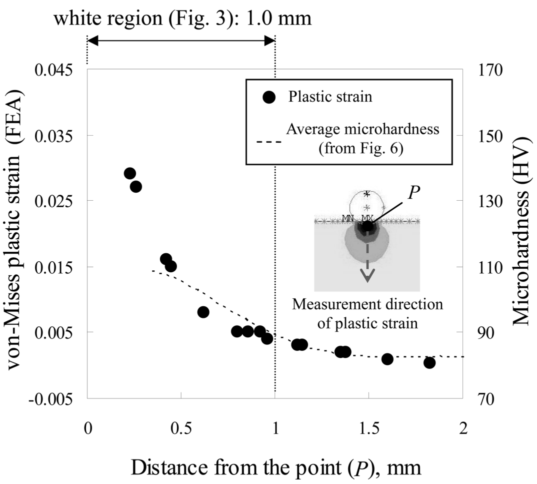

Fig. 8 von-Mises plastic strain measured from point *P*

attributed to the plastic deformation. This may suggest that the white area shown in Fig. 3 could also be affected by the plastic deformation zone (Okayasu *et al.* 2008, 2011). To verify this, microhardness measurements before the etching technique were performed. The hardness was measured radially from point *P*, where the sample was dented severely. Fig. 5 shows the directions of microhardness measurement indicated by the arrows, and the results obtained are given in Fig. 6. It is clear that the hardness near the dented zone is high compared to the rest of the region. The average of the hardness near point *P* is found to be 124% of that in the other area. Since the depth of the high hardness area, about 1.0 mm from the point *P*, is close to the width of the white area shown in Fig. 3, the brightly etched areas would be influenced by the work hardening. To further substantiate whether the white area in Fig. 3 is related to the plastic zone or not, FE analysis was conducted. Fig. 7 depicts the FE plastic strain distribution for the tough pitch copper calculated with

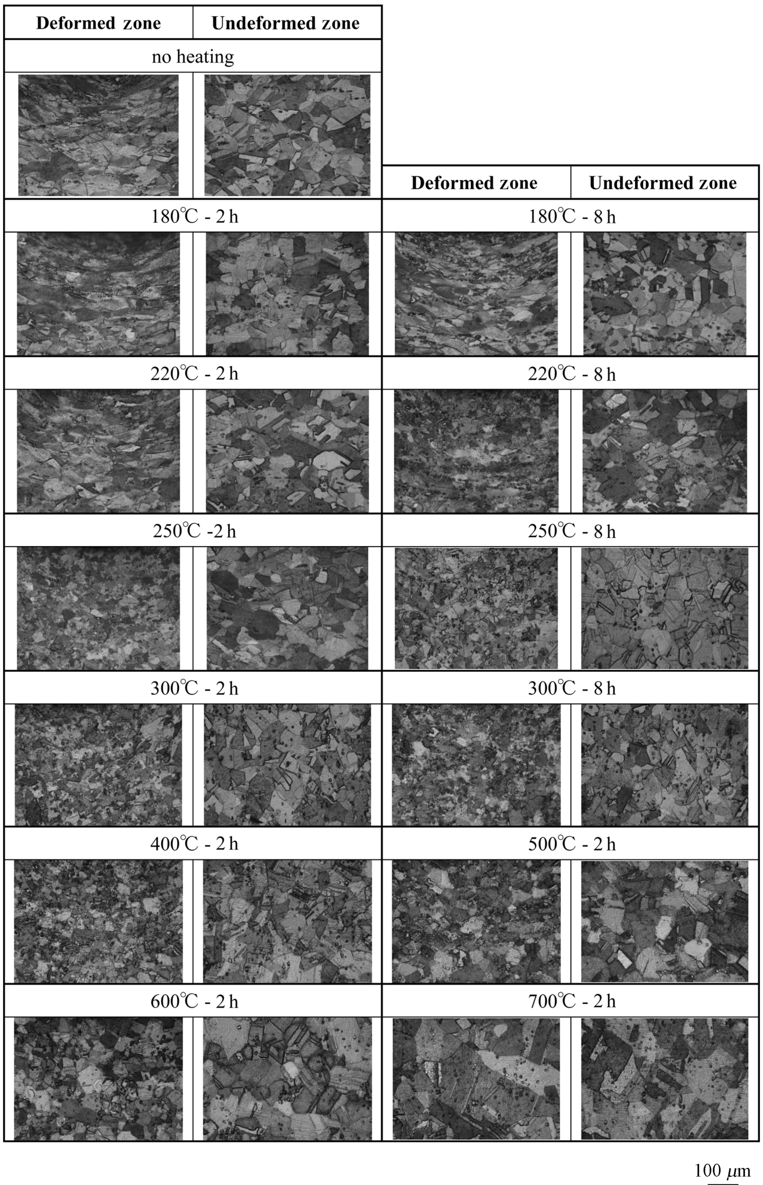

Fig. 9 Optical micrographs of the samples after etching

von-Mises criterion. It is observed that the plastic strain distribution, particularly shape and size, is similar to the bright region. In fact, the depth of the plastic strain region (more than 0.3) is about 1.2 mm. Hence, the bright area in the specimen stands for the actual plastic deformation zone. Fig. 8 displays the variation of plastic strain obtained by the above FE analysis. As seen the strain level is high in the sample adjacent to the contact point by the wire, whereas the hardness level is almost constant in the area about 1.0 mm far from the point *P*. The mean hardness in Fig. 6 is also indicated in Fig. 8 by the dashed line. It is clear that the variation of the mean hardness is similar to

(a) Eular maps (b) Crystal orientation maps

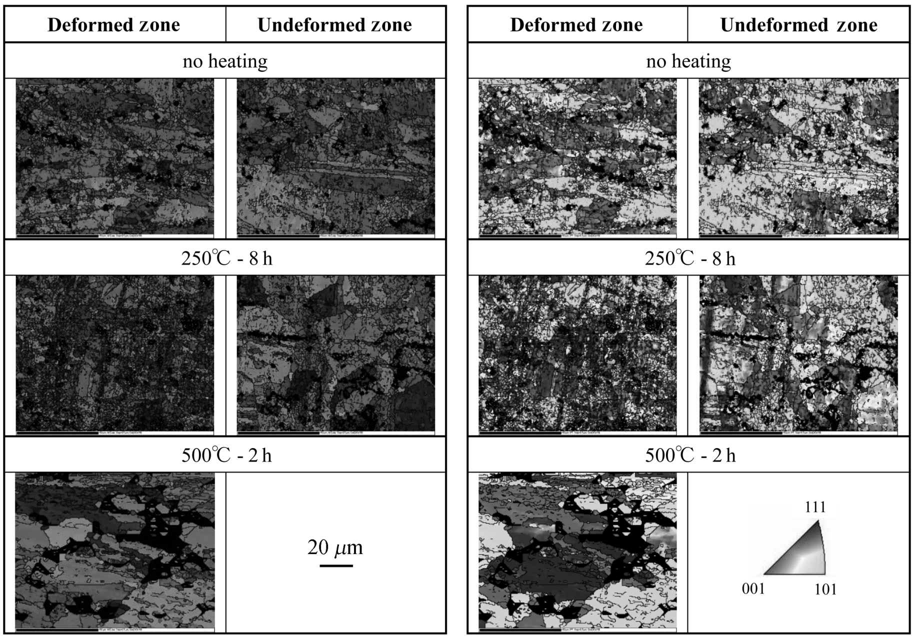

Fig. 10 (a) Eular maps and (b) crystal orientation maps for Samples A, G and K

the plastic strain results.

In order to interpret the mechanism behind the revealing of the plastic deformation zones, the microstructural characteristics of the tough pitch copper were investigated. Fig. 9 presents the microstructure of the undeformend and deformed zones for all samples. From the microstructural observations, three distinct microstructural characteristics were obtained in the deformation zone. More severe plastic deformation can be seen for the sample heated to below 220°C, which is similar to that for the sample before the heating process. Recrystallization occurs in the samples heated to 250°C-300°C (Sudo *et al*. 1997). Heating to high temperature (more than 500°C) promotes grain growth.

So as to understand the microstructural characteristics in detail, the crystal orientations were investigated by EBSD analysis. Figs. 10(a) and (b) depicts the Eular maps and the crystal orientation maps of Samples A, G and K. It should be noted first that the Eular map and crystal orientation map are obtained from the same area of each sample. Because of similar microstructural formation in the un- and deformed zone of Sample K, the analysis was conducted only in the deformed zone. Furthermore, the thin black solid lines (e.g. grain boundary) in the Eular maps and crystal orientation maps show high misorientation angle of more than 5°. As in the underformed zone of Sample A (no heating), randomly oriented crystal orientations are observed, and the misorientation angle in this sample seems to be relatively high compared to that in Sample K (500°C - 2 h). In this case, the high misorientation angle in the undeformed zone of Sample A may be affected by the rolling process to make the copper samples, and the low misorientation angle in Sample K could be influenced by the heating process, where the residual stress is reduced. Severe plastic flow is detected in the deformed zone of Sample A. Although heating process was carried

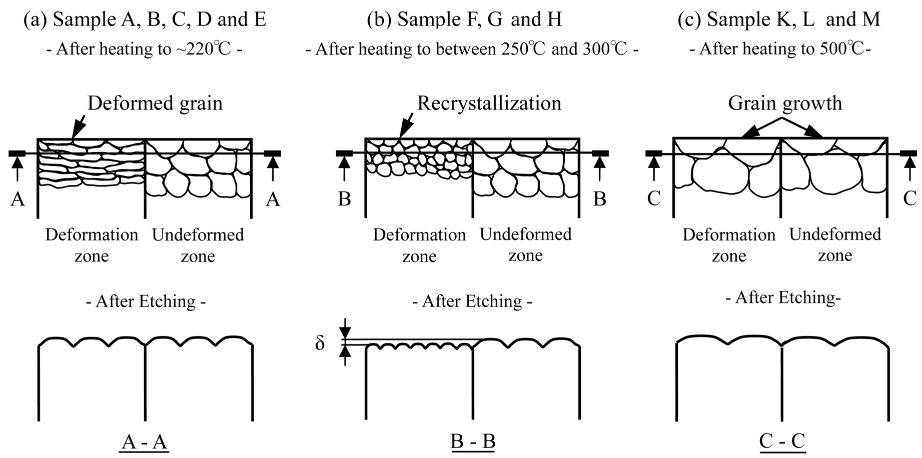

Fig. 11 Schematic illustration showing the change of microstructure and etching characteristic of tough pitch copper after heating to: (a) less than 220°C, (b) 250°C - 300°C and (c) more than 500°C

out in Sample G (250°C - 8 h), there is no clear microstructural difference in the undeformed zones, which is similar to that in the undeformed zone of Sample A. In the deformed zone in Sample G, the grains are collapsed and crystal orientation is different in tiny regions in each grain. From this analysis, it can be considered that the reason for revealing the plastic deformation zone (e.g. Sample G) is affected by the different microstructural characteristics.

Fig. 11 displays a schematic illustration of the three different microstructures and etching characteristics: (a) less than 220°C, (b) 250°C - 300°C and (c) more than 500°C. On the basis of the etching characteristics, the reason for the plastic deformation zone revealed in Samples F, G and H can be interpreted. Because the main corrosion occurs at the grain boundaries in the tough pitch copper (Fig. 4), a different severity of etching occurs between the undeformed zone and the recrystallized deformed area, with more severe etching in the deformation zone and only slight etching in the undefomed zone.

4. Conclusions

An etching technique for revealing the plastic deformation zone in a tough pitch copper has been studied experimentally. The following conclusions can be drawn.

(1) The localized plastic deformation in the tough pitch copper can be revealed using the proposed etching procedure. The process of the etching technique is as follows: (1) the specimens are plastically strained then heated to promote recrystallization (250°C - 300°C) before polishing to a mirror finish and (2) the polished surface is etched for 300 sec with an etchant consisting of 25 ml nitric acid solution and 75 ml water.

(2) The sample in the plastic deformation zone is etched severely while it is only slightly etched in the undeformed area. From the different severity of etching, the deformation zones become visible. The profile of the plastic deformation zone identified by the etching technique is in good agreement with the shapes of the work hardening zone and FEA plastic strain region.

(3) The plastically deformed zone, as revealed in the etching technique, is attributed to a change of

microstructure. The microstructure in the plastically deformed zone is altered by heating (250°C - 300°C), in which recrystallized small grains are created. Moreover, it is appeared by the EBSD analysis that the crystal orientation is complicated in tiny area in each grain for the samples heated to between 250°C - 300°C.

Acknowledgements

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