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AN IMPROVEMENT OF A TIP PREPARATION TECHNIQUE

H. Morikawa, K. Goto*, F. Iwatsu and T. Terao

Department of Materials Science and Engineering, Nagoya Institute of Technology,
Gokiso-cho, Showa-ku, Nagoya 466, Japan

*Department of Systems Engineering, Nagoya Institute of Technology,
Gokiso-cho, Showa-ku, Nagoya 466, Japan

Résumé - La méthode pour préparer les pointes de Mo par le polissage électrochimique a été améliorée en employant du courant alternatif à très basse fréquences. La relation entre les courants et les tensions a été obtenue et l'avancement du polissage a été discuté du point de vue de la formation et de l'enlèvement de la couche d'oxide sur la surface de la pointe.

Abstract - An improvement of a tip preparation technique of Mo by electrochemical polishing has been accomplished by using an alternating current of very low frequencies. The relationship between the currents and the voltages has been obtained and then the progress of the polishing is discussed from the view point of the formation and the removal of the oxide layer on the tip surface.

1. Introduction

The preparation of sharp-pointed tips is of basic importance for field ion microscopy. Sharp tips are also being required as an electron source in various instruments, e.g., a transmission electron microscope, TEM, a scanning tunneling microscope, STM, etc. Various techniques, therefore, have been developed, e.g., chemical or electrochemical polishing[1], ion sputtering[2], etc.

An improvement in a tip preparation technique of Mo has been accomplished by modifying an electrochemical polishing with A.C. The looks of the whole tip, the surface roughness, preferential polishing of a part of a tip near the surface of the solution, and the sharpness of the polished tip are found strongly influenced by the frequency, the voltage and the wave shape of alternating current (A.C.) applied. The I-V characteristics, i.e., the relationship of the current with the voltage applied, are also obtained in order to study the reaction of polishing. It is attempted to interpret the effects of the formation and removal of an oxide surface layer on the progress of the tip preparation.

2. Experimental

The A.C. polishing of Mo in an aqueous solution of 5% KOH is taken up first because of its simple system. In order to control the conditions of polishing, a function generator, a newly-built controller (H.Morikawa and K.Goto, in preparation) and a commercially available audio amplifier are used in series, Fig.1. The wave shapes, such as sinusoidal, triangular or rectangular, are generated

by the function generator. The phase angle of the wave and the number of waves to be applied are controlled by the controller. The applied voltage can be controlled either by the function generator or by the controller. The voltage and the current are monitored by an oscilloscope. A telescope is provided to observe the progress of the polishing. The changes of the tip shapes have been observed by an optical microscope.

3. Results

The polishings with sinusoidal, triangular or rectangular waves or the modified ones from them have been carried out. No remarkable difference in the results of polishing with these wave shapes has been recognized so far in the tip shapes, as long as they are compared at a same effective value of the voltage. In this study, therefore, triangular waves are almost exclusively employed because linearly increasing or decreasing voltage seems better for studying the relation between the reaction and the tip shapes obtained using the I-V characteristics. In the case of polishing by triangular waves, the polishing have progressed through various shapes depending on the applied voltages and the frequencies. The optical micrographs of tips on the midway of polishing are shown in the frequency-voltage diagram, Fig.2. The locations of x under each photographs indicate the polishing conditions, the voltage and the frequency. At very low frequencies lower than 1Hz, which are not a practical condition for tip making, the dependences on the applied voltages are not remarkable. At very high frequencies, sharp tips have not been obtained. Our attention, therefore, has been focused on frequencies in between 1Hz and 100Hz.

In order to reveal the reaction of polishing of molybdenum with A.C., I-V characteristics for various frequencies are taken by using the counter electrode of tungsten wire. These characteristics are found to depend on the frequencies applied, Fig.3. In the case of 1/4 Hz which is thought to correspond to a D.C. case, as the voltage

Fig. 1 The block diagram of the circuit for polishing.

Fig.2 Mo tips are polished in 5% aqueous KOH solutions with A.C. whose wave shape is triangular with various frequencies and various applied voltages.

is increased the current begins to increase at a threshold value around 2V, saturates at around 4V and then sharply decreases at 7-8V. Sometimes the current increases very steeply again at around 9V accompanied by strong bubbling around the tip, which means the progress of the electrolysis of water. On reducing the applied voltage after passing the maximum, the current shows again a peak at around 5V. In the negative potential side, the current vibrates violently because the electrolysis of water at the tip surface gives rise to strong bubbling. The peel off of a surface layer, probably an oxide layer grown under positive potential, can be seen with the telescope just after the bubbling starts when the tip potential has changed from positive to negative. The results of 1Hz, Fig.4(b), are almost similar to those of 1/4 Hz case, Fig.4(a). The saturation of the current observed in the low frequency cases, however, can not be seen when the frequency increases to 10 Hz, Fig.3(c). At 60 Hz, which is the frequency of the power commercially supplied, the result,

Fig. 3 The I-V characteristics in the case of polishing of a Mo tip with 5% aqueous KOH solution. The thick line represents the first cycle starting from 0V and increasing the potential to the positive side first. The thin line is of the second cycle following the first one.

Fig.3(d), resembles that of 10 Hz case. It is also noted that the characteristics of the negative sides seem not to remarkably depend on the frequencies.

4. Discussion

Three different features of tip shapes have been recognized as illustrated in Fig.4, i.e., (a) with a narrow part formed near the interface of the polishing solution and the air, (b) with a gradually tapered short shank and (c) with a thin and long shank. The feature (a) appears at voltages higher than certain voltages which increase gradually from around 7 V at 5 Hz to around 11 V at 60 Hz. This feature may come from high mobility of ions at the liquid surface. The feature of (c) appears when polished at low voltages and low frequencies. The tip end, however, is not sharp in this case. By contrast, the tip of (b) type seems to give a sharper tip end than of (c) type for tips with the same width of the shank parts.

Depending on the anodic dissolution mechanism in a basic solution proposed by Johnson et al.[3], molybdenum is oxidized to form Mo_2O_5 first, and then dissolves very slowly into aqueous solution by forming a molybdate ion, MoO_4^{--} . As the oxide grows on the tip surface, dissolution of the metal must become slow and eventually may almost cease when the thickness of the oxide reach a certain value. This also can be seen from the I-V characteristics for the low frequency, Fig.3(a). When a potential of the tip is kept positive for a long time, the oxide may grow to a certain thickness which is almost same everywhere on the tip surface, even though the dissolution speed usually depends on the curvature of the surface. For the higher frequency, Fig.3(c), such saturation of the current can not be seen because the voltage begins reducing before the completion of the protective oxide layer on the tip surface. The tip end, therefore, is dissolved faster than the shank area, resulting in the (b) type tips. In addition, the I-V characteristics of the first cycle apparantly differs from all the other cycles followed. The effect of the oxide formations described above is more conspicuous in the first cycle than in all the other cycles followed. The intermittent application of the A.C. voltage one cycle by one cycle, therefore, inclines to result in the (c) type polishing.

Fig. 4 Three typical features which are depicted and emphasized from the observed tip shapes ; (a) with a narrow part formed near the surface of the polishing solution, (b) with gradually tapered short shank and (c) with a thin and long shank.

The influence of bubbling of course can not be ignored. Especially in the case of polishing with high frequencies, continuous bubbling seems to give rise to the convection of the solution. The bubbling may sometime disturb the reaction of the tip with the solution, but on the other hand may sometime promote the reaction through the convection by supplying the new solution to the proximity of the tip surface.

Depending on the present results, the frequency of A.C. for the polishing of tip is one of the effective factors as well as the well known factors, e.g., the voltage, the movement of the tip during polishing, bubbling, the convection of the solution and so on. In order to make a fine tip, for instance, it seems a good idea to start polishing with the frequencies for the (b) type and then finish the tip end at the conditions for the (c) type.

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