RECENT EXPERIMENTS ON VACUUM BREAKDOWN OF OXYGEN-FREE COPPER ELECTRODES

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ABSTRACT

The effects of the gas content, *in situ* sputter cleaning, annealing and a mirror finish of oxygen-free copper (OFC) electrodes on the electrical breakdown in a vacuum were investigated. The grade of the OFC meets the ASTM-F-68 Class 1 standard. Electrochemical buffing (ECB) or diamond turning was employed for obtaining a mirror finish. It was found that every one of parameters investigated, namely, low gas content, *in situ* sputter cleaning and annealing, increases the breakdown fields after conditioning due to repetitive breakdowns. In addition, a higher annealing temperature produced higher breakdown fields. The mirror finish reduced the required number of breakdowns (conditioning procedure by repetitive breakdowns) to improve the insulating capability of the vacuum gap. A breakdown field of about 250 MV/m was obtained by a combination of annealing at 700 ℃ and a mirror finish by diamond turning. It has been pointed out that the combination of these treatments is necessary for obtaining higher breakdown fields. The effect of annealing and diamond turning is discussed on the basis of the recrystallization of copper and residual stresses on the surface.

1. INTRODUCTION

Particle accelerators, vacuum interrupters, and other such devices utilize the excellent insulating capability of a vacuum. In many cases, however, the performance and reliability of these devices are limited by the electrical breakdown in a vacuum.

It has been believed that electrical breakdown phenomena in a vacuum are greatly affected by the surface conditions of the electrodes. In electron storage rings, however, the electrodes are irradiated by high-energy photons that deeply penetrate into them. Accordingly, occluded gases, as well as adsorbed gases on the electrode surface, are desorbed from the electrodes [1]. These desorbed gases limit the electron storage time and cause electrical breakdowns in the accelerating tube.

The electrodes used in these kinds of devices should have low gas contents and high breakdown strength. It is generally recognized that those materials that have a high breakdown strength are the same as those used in ultra-high vacuum technology, that is, they should be gas-free [2]. The electrodes in these devices are, therefore, often made of oxygen-free copper (OFC).

In designing such devices, it is necessary to evaluate how the purity, especially the gas content, of the OFC electrodes affects their electrical breakdown characteristics. However, it is difficult to discriminate the effects of the bulk characteristics from those related to the surface condition, since electrical breakdown in a vacuum is largely determined by the electrode surface conditions.

Generally, electrode surfaces are polished or machined to be smooth [3], since one cause of the electrical breakdown of vacuum gaps is field emission from sharp protrusions on the cathode electrode. Polishing or machining, however, sometimes increases the residual stresses that tend to affect the field emission characteristics, and, thus, the breakdown strength. Electrochemical buffing (ECB), or diamond turning can be used not only to produce mirror finish, but also to reduce the residual stresses caused by polishing or machining. The annealing of bulk materials is also often performed to improve the breakdown strength, since annealing may has effects that reduce the gas contents and recrystallize the material to lower crystal strains.

This paper deals with recent investigations to evaluate the breakdown characteristics of OFC electrodes processed by bulk treatments and the surface finishes mentioned above.

2. OFC COPPER AND EXPERIMENTAL PROCEDURE

The electrodes were made of vacuum-degassed oxygen-free copper with a purity satisfying the ASTM-F-68 standard, and having a micro-structure ranging from Class 1 to Class 5. The degrees of contamination in the micro-structure are given in Fig. 1 [4]. The structure of Class 1 is the purest form of OFC, since its micro-

0-7803-3906-6/96/$5.00 ©1996 IEEE   XVIIth International Symposium on Discharges and Electrical Insulation in Vacuum, Berkeley 1996

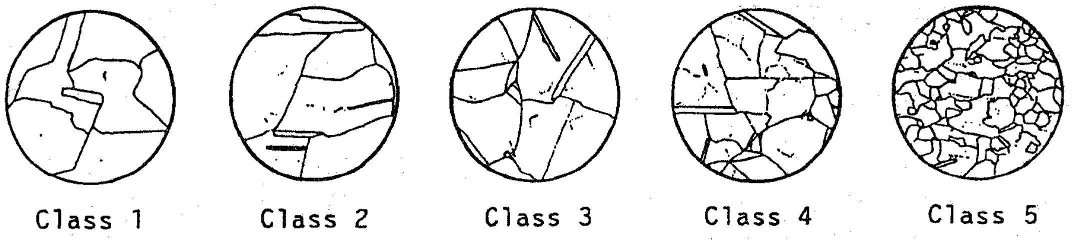

Fig. 1 Microstructure of different grades of oxygen-free copper.

structure has the fewest porosities. Since these porosities are formed by gases absorbed in molten copper, their density increases with the amount of dissolved gases. The minimum porosity therefore corresponds to the minimum gas content. The typical chemical composition of the OFC used was : for Class 1, >99.996 % Cu, <0.5 ppm hydrogen gas, and less than 2 to 3 ppm oxygen ; for Class 5, >99.996 % copper, <3 ppm hydrogen gas, and 5 to 8 ppm oxygen.

The electrodes were first roughly machined by turning them to a mushroom shape. Some of them were then annealed in a vacuum for one hour at 400 ℃ or 700 ℃. Some of the machined electrodes and some of these annealed electrodes were then machined by electrochemical buffing (ECB) [5] or diamond turning [6] to a mirror finish.

The experimental procedure was as follows. All of the electrodes were first ultrasonically cleaned in an acetone bath for 10 min. and then placed into a vacuum chamber, where their surfaces were cleaned by He or Ar ion bombardments. The electrodes were then moved to a vacuum chamber in which the electrode surface could be analyzed by X-ray Photoelectron Spectroscopy (XPS). Afterwards, a series of 500 breakdowns was carried out for each test gap. All of the processes could be carried out in a vacuum without any exposure of the electrodes to air (*in situ* experimental system). The applied voltages were positive impulses (64 μs rise and 700 μs decay) reaching a maximum of 100 kV. The pressure in the vacuum chamber before the breakdown tests was reduced by a sputter-ion pump and a Ti-getter pump to 1-3x10^{-8} Pa. The details concerning the experimental system have appeared elsewhere [7].

3. SURFACE CONDITIONS OF ELECTRODES BEFORE AND AFTER BREAKDOWNS

The electrode surface conditions analyzed by XPS before and after breakdowns are shown in Fig. 2. This figure clearly shows that the electrode surface before breakdowns is covered by a contaminant layer comprising oxides (O_{1S}) and hydrocarbon (C_{1S}). That is, at the beginning of the breakdown measurements, the observed breakdown voltage is the value of the contaminant layer other than the electrode material itself. On the other hand, after the breakdowns (in this case 500 times), the electrode surface is completely cleaned. That the peaks originate from the electrode material (Cu) has become clear, and no peaks due to contaminant elements, such as O_{1S} and C_{1S}, can be observed. This result means that repetitive breakdowns make the electrode surface clean, and, therefore, the cause of the conditioning effect achieved by repetitive breakdowns seems to be a cleaning of the electrode surface. As described in the next section, however, the cleaning of the electrode surface is not always a sufficient condition to obtain a higher breakdown strength, but a necessary condition.

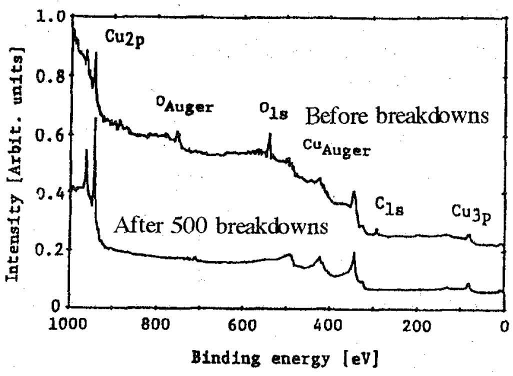

Fig. 2 XPS spectra of the cathode electrode surface conditions before and after 500 breakdowns

4. NECESSITY OF *in situ* SPUTTER CLEANING

Figure 3 shows the effects of cleaning using *in situ* Ar-ion beam-sputter cleaning (pre-cleaning)[8]. It is well known that the Ar-ion sputter cleans the electrode surface completely. Both pre-cleaning and non pre-cleaning were performed using Class 1 Cu electrodes. The effect of *in situ* sputter cleaning is obvious. For the pre-cleaned electrode, a significant conditioning effect

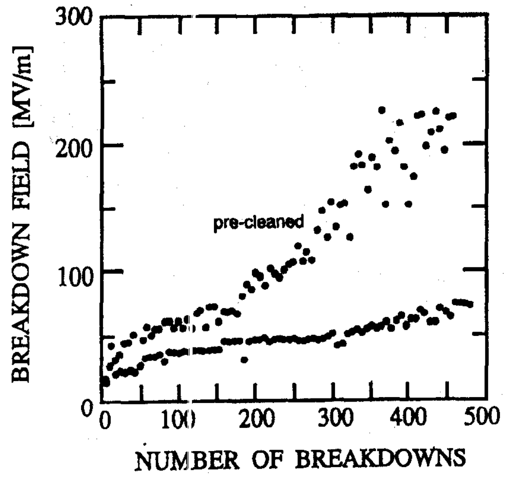

Fig. 3 Effect of *in situ* pre-cleaning performed by Ar-ion sputtering on the breakdown fields. Class 1 copper was used.

was observed, while for the non pre-cleaned electrodes, only a few improvements were made in the breakdown strength.

It should be noted that no significant differences existed in the breakdown fields between the pre-cleaned and non pre-cleaned electrodes at the first voltage application. According to Fig. 2, it can be expected that the breakdown field at the first voltage application is improved. In fact, however, no improvement was observed, though a distinct difference in the conditioning effect was observed.

This result reveals that *in situ* sputter cleaning is not effective for improving the breakdown strength at the first voltage application, but is necessary to clarify the effect of surface and bulk treatments on the breakdown characteristics and to obtain a better conditioning effect.

For all of the experiments described in this paper, therefore, the electrode surfaces were cleaned by an *in situ* sputter cleaning method.

5. BREAKDOWN CHARACTERISTICS OF CLASS 1 COPPER

The dependence of the breakdown strength (breakdown voltage/gap length) on the number of breakdowns is shown in Fig. 4. These results were obtained using Class 1 and Class 5 electrodes. Notice that the breakdown strength is improved with each breakdown, and that the rate of this improvement depends on the grade of the OFC.

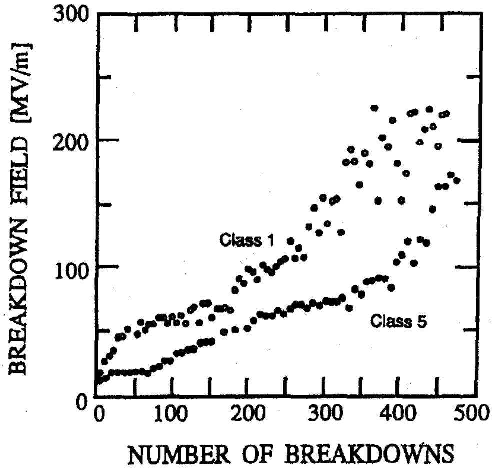

Fig. 4 Difference of the breakdown fields between Class 1 and Class 5 copper.

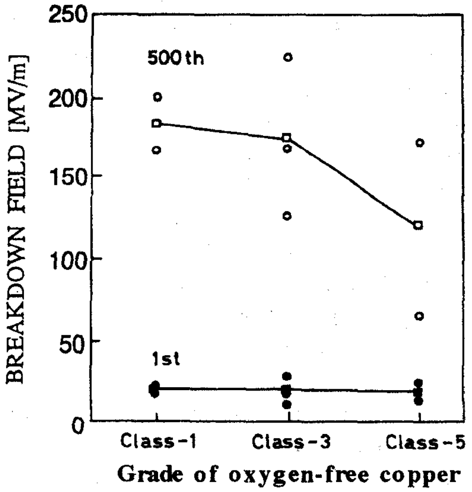

Fig. 5 Dependence of the breakdown fields on the OFC grade.

Figure 5 shows how the breakdown field strength depends on the grade of the OFC. The white circles represent the strength of the breakdown field at the 500th breakdown, and the solid black circles represent the breakdown field strength at the first voltage application. The rectangular marks denote the average value of measurements for each grade of OFC. These average values are listed in Table 1.

Table 1.
Breakdown field strengths for each grade of the OFC [MV/m]

Breakdown	Class 1	Class 3	Class 5
500th	183	173	118
1st	20	19	19

These results show that the breakdown-field strength depends on the purity of the electrode materials. High-purity electrodes, on average, show higher breakdown field strengths than lower purity electrodes, although the experimental data are scattered. In the following experiments, Class 1 copper is therefore used as the electrode material.

6. MIRROR FINISH BY ELECTROCHEMICAL BUFFING

Recently, an electrochemical buffing (ECB) technique has been developed to obtain ultra-smooth surfaces for use in extreme-high vacuum chambers, ultra-fine gas piping etc.[5]. Since this procedure makes the surface mirror-like, micro-protrusions that may cause field emission and increase the effective surfaces where gases are adsorbed can be removed. It may therefore be expected that the breakdown strength is improved.

Figure 6 shows the effect of ECB on the breakdown fields. There is a significant difference that the number of breakdowns necessary to achieve settled breakdown fields is greatly reduced. For electrodes processed by ECB, the number is about 40, while for only lathing it is only about 400. ECB polishing can reduce the residual stresses due to machining. This may contribute to a lower number necessary to achieve conditioned breakdown fields. Another cause of the reduction of the number may be the removal of protrusions. No significant differences have been observed in achieved breakdown fields for both electrodes. Also, the breakdown field at the first voltage application for the ECB-processed electrode is similar to that for lathing only the electrode, that is, 16 MV/m for the ECB electrode and 18 MV/m for the lathing only the electrode, respectively.

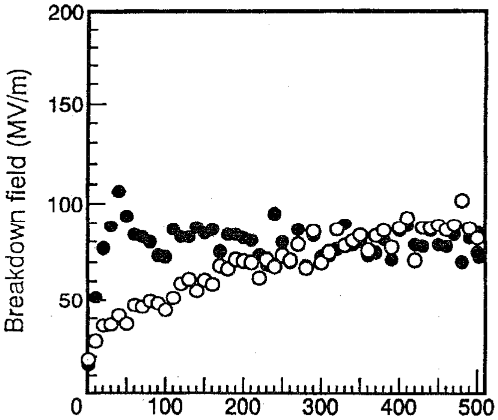

FIG. 6 Effect of ECB finishing on the breakdown fields. ●: ECB finish, ○: lathing only.

The important findings obtained from Fig. 6 are that protrusions on the electrode surfaces are not always the decisive factor for breakdown initiation, since the difference in the breakdown fields at the first voltage application is similar for both electrodes. That is, the removal of protrusions does not always contribute to an improved breakdown, if the electrode surface is carefully machined.

7. EFFECT OF ANNEALING THE ELECTRODE ON THE BREAKDOWN CHARACTERISTICS

As described in Table 1, it can be seen that OFC electrodes having fewer gas contents have a higher conditioning effect. The OFC used in our experiments was cast in a vacuum in order to lower the gas contents. The content of the absorbed gases can be controlled by changing the crystal structure. Thermal annealing was employed to change the crystal structure. Sample electrodes were heated at a temperature of 400 ℃ or 700 ℃ for 1 h in a vacuum. The vacuum chamber for the thermal annealing was separated from the vacuum breakdown measurement chamber. The electrodes after annealing were transferred from the annealing chamber to the breakdown measurement chamber through the air.

Figure 7 shows the effect of annealing on the conditioning effect. The breakdown fields gradually increase along with the accumulation of breakdowns, and achieve values corresponding to the annealing conditions. They then settle down to the final breakdown fields with a little scattering. Both the first and final breakdown fields are important values for representing the breakdown strength, since the difference between these two values shows the degree of the effect of conditioning.

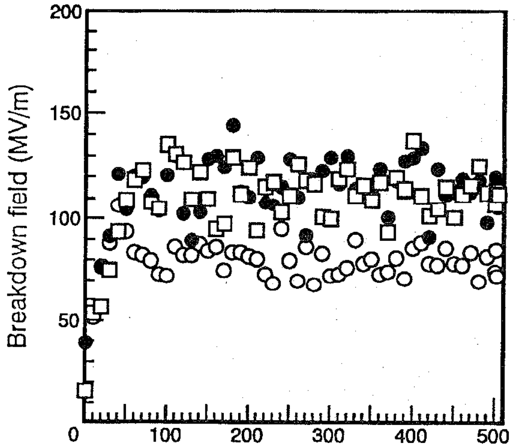

Fig. 7 Effect of annealing on the breakdown fields. The electrode surfaces were finished by ECB. □:annealed at 700 ℃, ●:annealed at 400 ℃ and ○:non annealed.

The values obtained from Fig. 6 are summarized in Table 2.

Table 2.
First and achieved breakdown fields showing the effect of annealing. The numbers in ( ) denote the range in which the breakdown fields were averaged.

	Breakdown fields [MV/m]	
Electrode treatments	1st	Achieved
Non annealed, ECB	15.6	80(100-500)
400 ℃, ECB	39.1	120(100-500)
700 ℃, ECB	16.0	115(100-500)

Table 2 explains that the achieved breakdown fields for annealed electrodes are higher than that for an unannealed electrode. The reason that the annealing affects the breakdown field may be that the annealing of electrodes reduces the gases contained in the electrodes, and releases any residual stress due to electrode machining.

8. DIAMOND TURNING AND RESIDUAL STRESSES

Diamond turning has been employed as another process to reduce the residual stresses caused by machining or polishing. Diamond turning also has a feature that produces a surface mirror. Figure 8 is a phase-contrast photomicrograph of the electrode surface after diamond turning. It shows that the pitch is 15 μm, and the surface roughness is 0.06 μm.

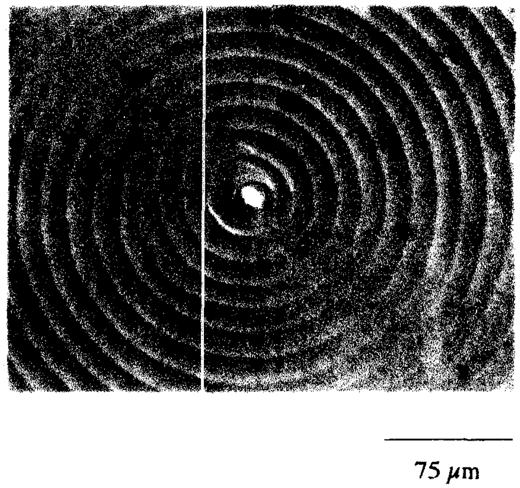

Fig. 8 Phase-contrast photomicrograph of diamond turning finished surface.

The residual stresses of the electrode surfaces were measured by an X-ray diffraction method. The results of the residual stresses before and after diamond turning are shown in Fig. 9. The white bars denote the residual stresses before diamond turning, while the black bars show the residual stresses after diamond turning in conjunction with the effect of annealing on the residual stresses. Before diamond turning the residual stresses scatter from negative (compressive stress) to positive (tensile stress) and their values are large. After diamond turning, on the other hand, all of the stresses become tensile stress, and their values are almost in the same range, but less scatter. It means that the mechanical structure of electrode surfaces finished by the diamond turning is almost in the same condition.

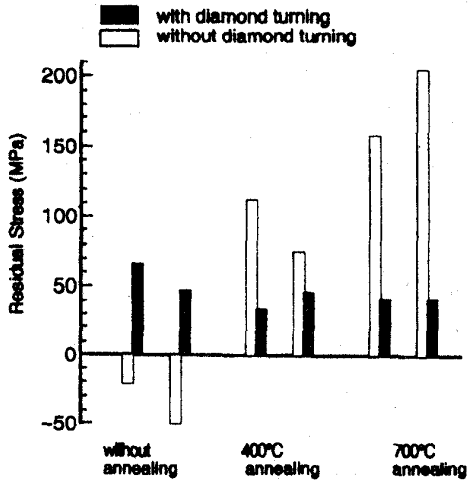

Fig. 9 Residual stresses before and after diamond turning in conjunction with the influence of annealing.

Three pairs of diamond-turning finished OFC electrodes were prepared:

Gap 1 : Only diamond turning.
Gap 2 : Annealed at 400 ℃ and diamond turning.
Gap 3 : Annealed at 700 ℃ and diamond turning.

The dependence of the breakdown field on the number of breakdowns is shown in Fig. 10. The

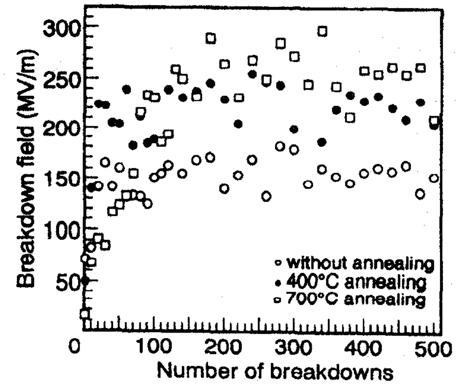

Fig. 10 Effect of diamond turning and annealin on the breakdown fields.

breakdown fields at the first voltage application are 71.4 MV/m for Gap 1, 49.4 MV/m for gap 2, and 14.9 MV/m for gap 3. As a result of the conditioning effect, these values gradually increase as the number of breakdowns increases, and then settle down to the final breakdown fields. The first and settled-down values obtained from Fig. 10 are summarized in Table 3. This table shows that the combination of annealing and diamond turning is an effective procedure for improving the breakdown strength. For that combination the breakdown field after conditioning reaches 250 MV/m.

Table 3.
The first and the average breakdown fields. The numbers in ( ) denote the range of the breakdown fields averaged.

	Breakdown field [MV/m]	
Samples	1st	Average
Gap 1	71.4	160 (50-500)
Gap 2	49.4	230 (50-500)
Gap 3	14.9	250 (150-500)

By comparing the values in Table 3 with those obtained in another experiment carried out with electrodes machined by ordinary turning [9], it was confirmed that diamond turning results in higher breakdown fields.

Furthermore, diamond turning results in a different conditioning effect. The conditioning effect shown in Fig. 10 is no longer apparent after about 100 breakdowns. A similar trend is seen in the result of stainless-steel electrodes polished with ECB [10]. Either diamond turning or ECB makes the electrode surface mirror-like, thus, greatly reducing the size and number of small protrusions, and, thus, the amount of gas-adsorbing sites and field-emission sites. It is therefore expected that the conditioning of vacuum gaps can be achieved with fewer breakdowns.

From the values listed in Table 3, we can see that the final breakdown fields of Gap 2 and Gap 3 (annealed) are higher than those of Gap 1 (unannealed). This may be because annealing reduces the gas content, and changes the quality of the electrode surface (recrystallization). The breakdown strength can thus be increased by annealing electrodes in a vacuum before diamond turning.

9. CONTROLLING THE CRYSTAL STRUCTURE BY ANNEALING AND BREAKDOWN CHARACTERISTICS

As described above, those Class 1 electrodes finished by diamond turning have a higher breakdown strength and a significant conditioning effect with raise at annealing temperatures of 400 ℃ to 700 ℃. To clarify that this effect can be attributed to whether the change of gas contents or crystal structure due to heating, it is necessary to control the crystal structure under the same annealing temperature.

To separate these two parameters (gas contents and crystal structure), electrodes processed in the following way were prepared. Firstly, copper materials having a cold reduction rate of 20 % and 50 % were prepared. These two copper materials have different recrystallization temperatures. Figure 11 shows the relationship between the annealing temperature of the OFC used in this experiment and the Vickers hardness (Hv). This figure demonstrates that the different crystal structures can be made under the same annealing temperature, since the recrystallization temperature is dependent on the cold reduction rate of the material. According to this figure, we then prepared four types of OFC electrode pairs : electrode pairs A and B were annealed in vacuum for 30 min at 310 ℃ to recrystallize the pair A, while pair B was not recrystallized, and electrode pairs C and D were not annealed, but their reduction rates were different.

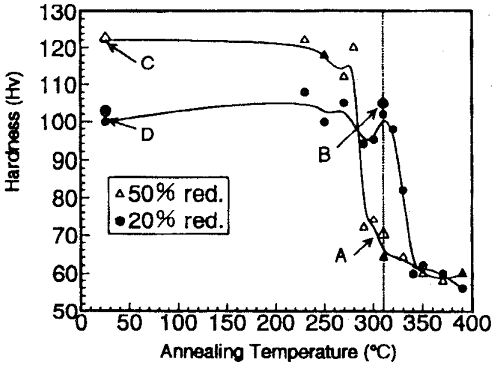

Fig. 11 Isochronical annealing curves of oxygen-free copper with 20 % and 50 % reduction.

The breakdown fields of these electrodes are shown in Figs. 12 and 13. Figure 12 shows the effect of annealing ; that is, electrode pair B was annealed at 310 ℃, while electrode pair D was not annealed. These two electrode pairs have the same Vickers hardness, and, thus, the same crystal structure. For both electrode pairs conditioning has been achieved after around 150 breakdowns ; then, the breakdown fields settled down to the values for each electrode pairs. Taking these settled values as their own breakdown field, it is obvious that the breakdown field for B is higher than that for D. This means that the reduction in the gas contents due to annealing caused an improvement in the breakdown field, since both electrode pairs were not recrystallized.

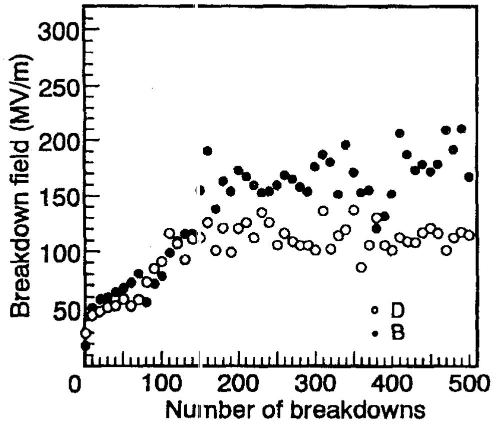

Fig. 12 Breakdown properties of electrode B (heat treated) and D (non heat treated) that are machined from a 20 % reduction.

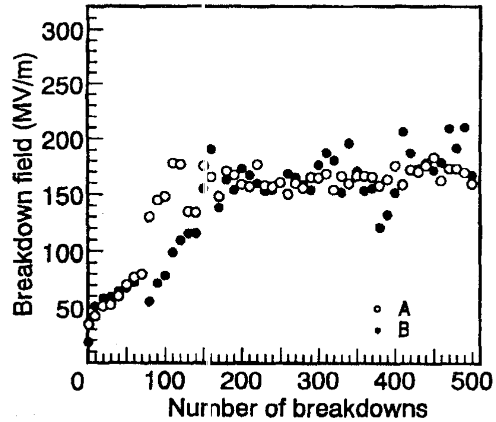

Fig. 13 Breakdown properties of electrode A (after recrystallization) and B (before recrystallization) that were heated at the same temperature.

On the other hand, Fig. 13 compares the effect of recrystallization. For electrode pair A the residual stresses caused by machining were released by heating ; also, the size of the crystal grains was minimized by the recrystallization. It can be seen that conditioning was achieved after about 150 breakdowns. Moreover, the settled breakdown fields are almost the same for both electrode pairs, and the value is about 170 MV/m. From these two figures, it can therefore be concluded that a reduction in the gas contents due to annealing is more effective on the breakdown strength. In addition, no effect of hardness could be observed, since the breakdown strength for the electrode pair C was almost the same as that for electrode pair D. It seems that the hardness or the recrystallization does not influence the breakdown strength. However, it should be noted that the Vickers hardness denotes the value at the several 10 μm layer beneath the surface, where little effect on breakdown properties is expected.

The measured residual stresses on the electrode surface are shown in Fig. 14. The values for the anode and cathodes are shown separately for each electrode pair. The residual stresses were estimated by measuring the diffraction line emitted from the region irradiated by X-rays collimated by a 4 mm x 2 mm slit. The estimated values indicate the stresses in the region of 2-4 μm beneath the surface. Before breakdown measurements, that is, after diamond turning, the residual stresses have low value for almost all of the electrodes. This means that diamond turning releases the distortion of machined layers. Therefore, the distortions of the surface layers (2-4 μm) after diamond turning are almost even for each electrode.

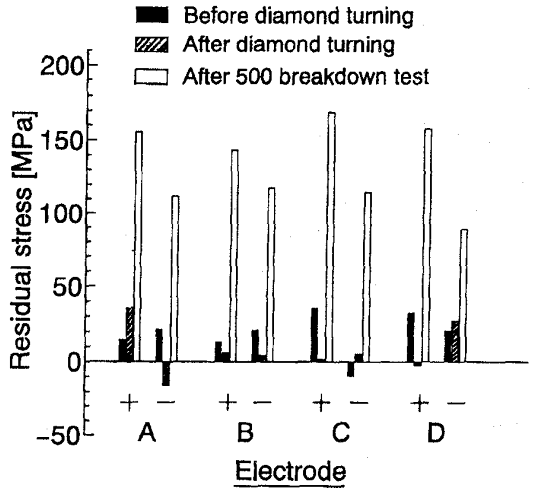

Fig. 14 Residual stresses of oxygen-free copper electrodes before and after diamond turning and after 500 breakdowns.

All of the residual stresses of the electrodes after breakdown measurements become positive (tensile direction) ; these values become significantly larger than those before breakdown measurements. Traces of melting of the electrode surfaces were observed after breakdown measurements [11]. The increase in the residual stresses after breakdowns can therefore be attributed to tensile distortion occurring during solidification under the application of an electric field. In addition, it can be seen that the residual stresses of the anodes after breakdowns are larger than those of the cathodes. This may be due to heating by the electrons from the cathode.

Figure 14 strongly suggests that the recrystallization or the stress influences the breakdown strength. However, the dependence of the Vickers hardness, which is subject to the crystal structure, was not observed. As described above, the layer where the Vickers hardness is measured is too deep to give effects on the breakdown strength. In order to reveal the effect of the crystal structure more in detail, it is necessary to analyze the depth profile of hardness.

10. SUMMARY

The experimental results obtained by using OFC electrodes were reviewed. The important findings can be summarized as follows :

(1) Low gas content copper, that is, vacuum-degassed Class 1 copper has a higher hold-off voltage capability.
(2) To obtain a better breakdown strength, *in situ* surface cleaning is necessary.
(3) The annealing is effective in improving the breakdown strength.
(4) A mirror finish by the electrochemical buffing or diamond turning reduces the required number of breakdowns to achieve a higher breakdown field.
(5) The combination of annealing and diamond turning is effective to obtain a high breakdown field ; the field reaches about 250 MV/m.
(6) The effect of diamond turning was discussed on the basis of the residual stresses on the electrode surfaces.

11. ACKNOWLEDGMENTS

This paper is the result of the long collaboration between National Laboratory for High Energy Physics (KEK) and Saitama University.

The author is particularly indebted to Prof. Y. Saito in KEK for his continuing interest and encouragement.

The author wishes to thank Mr. T. Sugano and Mr. K. Takeuchi of Mechanical Engineering Laboratory for their support for diamond turning.

Also, thanks are due to Mr. Y. Baba and Mr. K. Sato of Ultra Finish Technology for obtaining mirror finish by electrochemical buffing.

Finally. the author is grateful to Mr. Y. Nagai of Hitachi Cable Company for preparing oxygen-free copper electrodes.

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