Field Emission Current from High-$T_c$ Ceramic Cathodes

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ABSTRACT

In this work the result of emission phenomena investigations are presented. Experiments were conducted for high-$T_c$ superconducting ceramics YBaCuO and BiPbSrCaCuO. Fowler-Nordheim characteristics and the current-temperature dependencies were observed for superconducting and non-superconducting cathodes. It was noticed that the emission current value abruptly changed in the vicinity of the critical temperature. All measurements were done in a vacuum chamber at $10^{-6}$ Pa. X-ray diffraction analysis and scanning electron microscopy examination of the ceramic electrodes were performed also.

1 INTRODUCTION

Densities of the critical transport current are not high enough for applications of high-$T_c$ ceramic superconductors in electric equipment, i.e., in cables, generators, and transformers. The possible applications are in particle accelerators, cavity resonators, and fast superconducting switches. For the above mentioned purposes it is necessary to evaluate the performance of the superconducting ceramics under high electric field and strong vacuum treatment conditions and to investigate field emission phenomena as well as the emission characteristics of the materials.

2 EXPERIMENT

2.1 PREPARATION OF THE CATHODES

YBaCuO and BiPbSrCaCuO ceramic cathodes were made employing sintering in the solid state [1]. For the YBaCuO samples the temperature treatment was applied at 980°C and then they were conditioned at 620°C in order to increase the amount of the oxygen in the composite. BiPbSrCaCuO cathodes were sintered at 800°C and after that conditioned at 840°C [2]. All substrates that had been used were of technical purity. Specimens had the shape of discs 20 mm in diameter and were 2 to 3 mm thick.

Superconducting properties, the critical temperature $T_c$ and the critical current density $j_c$ of the cathodes were measured in a standard setup. All samples were ultrasonically cleaned in CCl$_4$ and ethanol directly before vacuum measurement. Standard surface cleaning methods employed for metallic electrodes treatment, i.e., heating in vacuum and argon plasma cleaning, turned out not to be applicable for these ceramic superconducting materials. The processes mentioned lead to a release of weak-bonded oxygen atoms from the crystal lattice and therefore cause material chemical degradation and disappearance of the superconducting phase.

2.2 MEASUREMENT SETUP

Selected samples were mounted in the measurement chamber. The vacuum system provided a pressure of $10^{-7}$ Pa without specimens. After cathodes were placed inside the chamber, the pressure level rose to $10^{-6}$ Pa. The measurement setup allowed external cooling of the cathodes fixed to the 'cold finger', using a silver binder and their temperature was monitored using a copper-constantan thermocouple. That thermocouple was in intimate contact with the sample. The cooling system made it possible to change the cathode temperature slowly from 77 K to the temperature of the surrounding environment. The distance between the cathode and the transparent anode [3], made from glass coated with a thin film of the InSnO composition, was controlled with a precision of 0.01 mm. All measurements were conducted using a 50 kV dc stable power supply and a high-resolution electrometer. Figure 1 presents the schematic measurement setup diagram.

3 RESULTS AND DISCUSSION

All measurements were done on emission originating from a single emission center on the cathode. This had been ensured by a visual observation through the transparent anode for currents $>10^{-8}$ A.

3.1 FOWLER-NORDHEIM CHARACTERISTICS

Current-voltage characteristics were presented in two sets of coordinates as $\log(I) = f(U)$ and Fowler-Nordheim (FN) $\log(I/U^2) = f(1/U)$. The typical current-voltage dependence for a superconducting ceramic cathode is shown in Figure 2. The measurement was done at two temperatures: 77 and 293 K, room temperature. The voltage was
gradually increased until the emission current was saturated. Then the voltage was decreased. The hysteresis effect in the current plot was observed at both temperatures. The saturation voltage and the switch-on voltage were higher at 77 K. The $F_N$ characteristics are linear within a certain range of the voltage only. Their slope indicates that the enhancement coefficient of the electric field at the emission center changes. The same behavior was observed for all superconducting samples, regardless their composition or superconducting parameters.

The same kind of measurement was done for a YBaCuO cathode that was not superconducting at liquid nitrogen temperature. That sample was of identical composition as the superconducting specimens, but contained less oxygen. It also displayed a current-voltage characteristic similar to that obtained for superconducting ceramics. The $F_N$ characteristics that were obtained in the experiment are similar to those that can be found in the literature for metallic cathodes, i.e. copper that is not a superconducting material, coated with thin dielectric layers [4,5]. It might indicate that the emission phenomena that took place for superconducting and non-superconducting YBaCuO and BiPbSrCaCuO ceramic cathodes not necessarily had to be influenced by the superconducting state. The origin of a flat part of the current record on the $F_N$ characteristic (Figure 2), which is nearly constant with voltage before the emission starts, is not sufficiently known. It is probably an ion current generated by partly ionized residual gases from the vacuum chamber and compounds desorbed from the sample surface.

### 3.2 CURRENT vs. TEMPERATURE DEPENDENCIES

The emissions current vs. temperature measurements were the second part of the experiment. Cathodes were cooled to 77 K. The voltage was fixed to obtain $\sim 1$ to $2$ nA of emission current, ensuring that the emission current density was lower than the critical current density needed for the phase transition from the superconducting to the normal state. Cathodes were then slowly heated up and the emission current was recorded. All superconducting samples displayed an abrupt increase of the emission current values when the temperature crossed the critical temperature of the cathode material. Figure 3 presents the typical emission current vs. temperature dependence. The resistance vs. temperature characteristic is also shown in order to illustrate that the jump in the current values takes place simultaneously with a superconducting phase transition. The emission current is stable below $T_c$ and becomes erratic and oscillates above that value.

The current vs. temperature measurement was carried out also for the non-superconducting YBaCuO cathode (Figure 4). In that case the abrupt change of the emission current was not observed. The emission current values increase smoothly with temperature.

After several sequences of measurements that had been done for the same superconducting cathode it was found that the effect of the abrupt jump of the emission current vanished (Figure 5). It was observed that after four measurements the current-temperature dependence became similar to the characteristic obtained for the non-superconducting samples.

The jump of the emission current in the vicinity of the critical temperature $T_c$ changes between consecutive measurement cycles, suggesting that the most important factor influencing the field emission is the gas layer present on the cathode surface.

The role of that insulating layer is not sufficiently explained. There are several models that describe the emission phenomena occurring on the insulator-coated cathode surface [6]. The most convincing model seems to be the Latham and Mazurek (LM) model. It assumes that
the cathode can be represented by the electrode-insulator-conductor-vacuum sandwich system. In this particular case it would be the system of the superconductor, a dielectric layer resulting from partial vacuum deoxygenation of its outer planes, a gas layer and conducting microparticles. Such microconductors may come from powdery intergranular material present in ceramic samples (Figure 6) or some particles left after polishing. It should be stressed that the surface of ceramic superconducting samples cannot be conditioned by heating and ion plasma cleaning, and therefore cannot be identified as absolutely smooth and clear.

Those microparticles may possess miscellaneous electrical properties and do not necessarily have to be superconducting. Initially, the conducting particle is separated from the surface of the cathode by the gas layer but when the voltage is applied the particle acts like an antenna and can cause a local electric field increase. That enhanced electric field can initiate the development of a conducting channel through the dielectric layer. If the channel is situated below the particle edge it is possible for some electrons to escape from it into the vacuum. Some electrons may be transferred to the particle and take part in a secondary emission from its surface. In this way an initiation of the emission center can be explained.

This model also describes successfully the existence of the jump in the emission current that occurs simultaneously with the change of the cathode temperature. There are no energy losses in the emission center below the critical temperature $T_c$. The emission current density at the emitter in that case should not exceed the critical value $j_c$, leading to a phase transition to the resistive state. The dielectric gas layer remains on the surface of the cathode. But when the temperature grows to a value $> T_c$, the emission center loses its superconducting properties, starts to be heated and the gas layer at that point is being desorbed. That liberated amount of gas is being ionized. The space charge created in that way disturbs the local electric field and causes oscillations of the emission current. The consecutive measurement cycles (Figure 4) confirm that the effect of the jump of the emission current is influenced by the desorption phenomena on the cathode surface.

### 3.3 MICROSCOPIC AND X-RAY ANALYSIS

An X-ray microprobe analysis and a scanning electron microscopy (SEM) microscopic examination of the cathodes were done before and after measurements of the emission currents. Figure 6 presents the surface of the cracked YBaCuO sample. It is clearly visible that there is fine intergranular material present among large crystal grains. SEM observation of the sample surface also reveals many low-dimensional irregularities, geometrical as well as chemical, which may be very efficient electron field emission centers. It should be stressed that the sample surface cannot be considered homogeneous and there may be microregions having dielectric properties. It is particularly important when we try to analyze emission phenomena.

Figure 7 is an example SEM photograph of the emission center for which the emission current density crossed the critical current density value $j_c$. Under such conditions the material loses its superconducting properties over a limited area and becomes resistive. The emission current starts heating the emitting zone, oxygen atoms are being liberated from the lattice, giving rise to the charge concentration in the direct emitter vicinity and a local thermal decomposition leading to dielectric by-products begins. When the local temperature is high enough, the
Figure 5. Four measurement cycles carried out for YBaCuO ceramic cathode. $T_1 = 87$ K, $\Delta T = 7$ K.

Figure 6. SEM image of YBaCuO sample fracture.

Emission point evaporates and the chemical composition in that place changes dramatically. Figure 8 shows the X-ray microanalysis result of the emission center before and after evaporation. It can be observed that concentration of copper and bismuth decreased significantly. It also indicates that the temperature of the emission point had been very high. Local increase in charge density as well as products of the evaporation may be breakdown dependent. It seems that a breakdown occurring at the emission point is a secondary phenomenon induced by a local phase transition between the superconducting and normal state.

4 CONCLUSIONS

Measurements of the field emission current from superconducting YBaCuO and BiPbSrCaCuO ceramics as a function of temperature revealed changes of the emission characteristics similar in nature to the ones observed in metallic electrodes. Therefore field emission phenomena in ceramic superconductors should rather be related to the solid state sorption phenomena than to a theory of superconductivity. Promising results have been obtained for very low emission currents. It was determined that there occurs a significant jump in the emission current character at the superconducting transition temperature. In the superconducting state low but very stable values are recorded while
in the normal state measurement data become unstable and erratic, increasing their value at several times.

REFERENCES


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