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Electrical electrodes of Ni-Me (Me=Ag, Mo, Cu) on $YBa_2Cu_3O_x$ surface

***Abstract**. The quality of metallic electrodes affects the measured critical temperature T_c and critical current I_c of high-temperature ceramic superconductors. In this paper the $YBa_2Cu_3O_x$ system with thin metallic film mono- and bi-layer conductors as a current and voltage electrodes is described. The influence of contact layer systems Ni-Cu, Ni-Ag and Ni-Mo on critical temperature and current density was examined. The simple model of physical phenomena in superconductor-metal interlayer has been presented.*

Streszczenie.** Jakość wykonania metalicznych elektrod ma istoty wpływ na mierzone wartości temperatury krytycznej T_c i prądu krytycznego I_c ceramicznych nadprzewodników wysokotemperaturowych. W pracy przedstawiono wyniki badań elektrod dwuwarstwowych. Elektrody te wykonano w celu poprawienia właściwości elektrod niklowych. Zastosowano rozpylanie magnetronowe z odpowiednich targetów w połączeniu z parowaniem próżniowym. **(Kontakty elektryczne z Ni-Me (Me=Ag, Mo, Cu) na powierzchni $YBa_2Cu_3O_x$).

Keywords: superconductor; resistivity; metallic electrodes.
Słowa kluczowe: nadprzewodnik; rezystywność; metaliczne elektrody.

Introduction

The main technological problem for high-current applications of ceramic superconductors is to prepare the metallic contacts on the surface of superconducting material. These contacts being the current electrodes should be characterized by very good mechanical properties and low resistance. Such features allow to avoid a drop of a basic superconducting parameters, such as critical temperature T_c and critical current density j_c. High-quality electrodes also enable the development of high-current practical applications of a new ceramic super-conducting materials.

There are many requirements for current (and voltage) electrodes deposited on surface of ceramic high-temperature superconductor. The resistance of electrical contact should be as low as possible to avoid the heat generation during high currents flow. The thermal resistivity has to be very low at the boundary, thus any heat generated will be quickly removed by the liquid nitrogen (coolant) - emergence and propagation of normal (non-zero resistance) zone in the superconductor volume will be unlikely.

Therefore the best materials for electrodes seem to be noble metals: gold, silver or platinum [1-2]. They have very low resistance yet in the form of thin film and they are resistant to environment conditions (changes of temperature, ambient atmosphere etc.). Unfortunately, there are some problems associated with them: atoms of these metals migrate from surface into the interior of porous granular superconductor very easily at elevated temperatures. Additionally, platinum can change the chemical and microstructural properties of ceramic material and thus it influences on its superconducting properties [3].

Other metals used as electrodes, such as copper, aluminium, indium etc. are usually unstable or react with ceramic substrate. Their electrical properties also are not good enough for electrical applications.

The method of electrode deposition should meet certain conditions (also dependent on the materials). First, it may not change properties of superconductor even locally. This is particularly important in the case of $YBa_2Cu_3O_x$–type superconductors because of the "mobile" oxygen. Of course, the used deposition technique can not be too complicated as well and it should give reproducible results. In some cases, it should allow to form on samples contacts of any shape, not only flat.

In practice several techniques are used for electrodes forming on ceramic superconductor surface. Among them may be mentioned: thermal vacuum deposition, vacuum deposition by electron beam using, cathode sputtering, magnetron sputtering, screen printing (thick films), metallic paste painting and much more.

In the previous works we tried to use nickel deposition as electrical contact on the $YBa_2Cu_3O_x$ superconducting samples [4]. Nickel was chosen as electrode material because it is used both in some electronic applications and for superconducting tapes production [5-7]. Unfortunately, that attempt was unsuccessful. Nickel layer was the heater, so it is not good electrode material. In the presented paper some modifications of Ni-$YBa_2Cu_3O_x$ compound and their properties are described.

Deposition of electrodes

Superconducting $YBa_2Cu_3O_x$ samples were prepared by standard solid state reaction from Y_2O_3, $BaCO_3$ and CuO substrates. They were characterised by critical temperature T_c of about 90 K and low values of critical current density j_c below 1 Acm^{-2}.

Nickel-metal electrodes were formed on ceramic superconductor surface to improve the electrical properties of pure nickel contacts. Metallic films were deposited on $YBa_2Cu_3O_x$ superconductor samples by magnetron sputtering from proper targets and by vacuum evaporation.

The procedure of deposition was as follows:

1) the substrates ($YBa_2Cu_3O_x$) were thoroughly washed in ethyl alcohol with ultrasound washer for ten minutes, dried in furnace at 150^{o}C for one hour, heated in vacuum of 2-4x10^{-3} Pa at 200^{o}C for a half an hour,

2) the Ni-Ag, Ni-Mo and Ni-cu contacts were formed on superconductor surface by method proper for contact material:

- Cu-Ni: Cu was vacuum evaporated for 3 min from tungsten boat, Ni was magnetron sputtered for 18 min,
- Ag-Ni: procedure as above,
- Mo-Ni: Mo was magnetron sputtered for 10 min, Ni – for 18 min,

3) current and voltage leads were attached to the metal films prepared on superconductor surfaces by soldering. This process caused some difficulties. After many attempts glycerine was used as a proper solder flux.

During process of magnetron sputtering the temperature of substrates (superconducting materials) was about 420-450 K.

Results of experiments

Superconducting transport parameters T_c and j_c for $YBa_2Cu_3O_x$ samples were measured by standard four-probe method. For this purpose the good low-resistance silver electrode was used. Value of T_c was typical for these materials (above 90 K). Also, the j_c values were like for bulk sintered granular oxide superconductors: tens of $mAcm^{-2}$. Additionally magnetic susceptibility measurements as a function of temperature were carried out and they showed strong diamagnetic Meissner effect. This indicated that the sample was superconducting in its volume and the current limit was caused by poor connections between the grains. However, such low j_c values were not a problem for contact resistance measurements.

To determine the metal/superconductor contact resistance three- and four-probe geometry was applied as is shown in Fig. 1. In such systems, the $U(I)$ characteristics were measured at room temperature and at liquid nitrogen temperature. On their basis the $R^{3\text{-probe}}(I)$ and $R^{4\text{-probe}}(I)$ dependencies were determined and then contact resistance $R_c(I)=R^{3\text{-probe}}(I)-R^{4\text{-probe}}(I)$.

The results of measured electrical resistance for three types of prepared electrodes are shown in Fig. 2-3 (Ni-Ag), Fig. 4-5 (Ni-Mo) and Fig. 6-7 (Ni-Cu).

For each kind of sample contact the dependencies of resistance of metal-superconductor interlayer on current flown value are shown. These relationships are presented both at room temperature and at boiled liquid nitrogen temperature.

As can be seen from Fig. 2-7 the values of contact resistance R_c are relatively large for each of the tested samples. This is observed both at room temperature and at the temperature of liquid nitrogen.

Moreover, in each case the contact resistance is lower at room temperature than at 77 K and this difference is very significant, particularly for Ni-Ag layer. Probably this indicates that between the electrode material and the superconductor a buffer area of semiconductor is created. This is possible because the substrate (superconductor surface) was not additionally cooled during sputtering process.

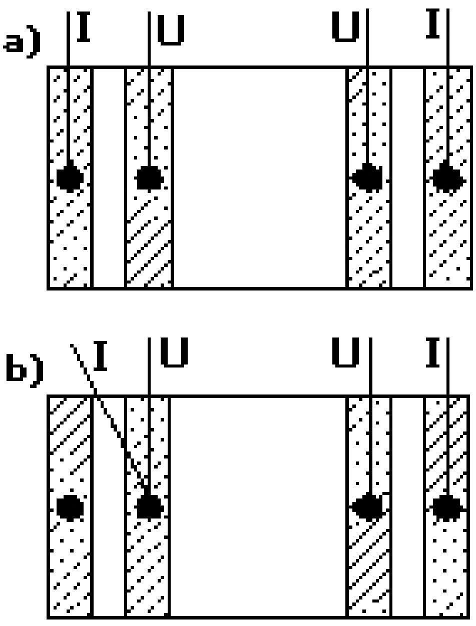

Fig.1. Geometry of contact pads on sample surface for a) four- and b) three-probe $U(I)$ characteristic measurements

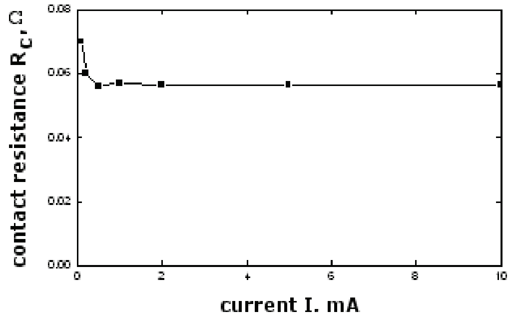

Fig.2. R_c of Ni-Ag interlayer at the room temperature

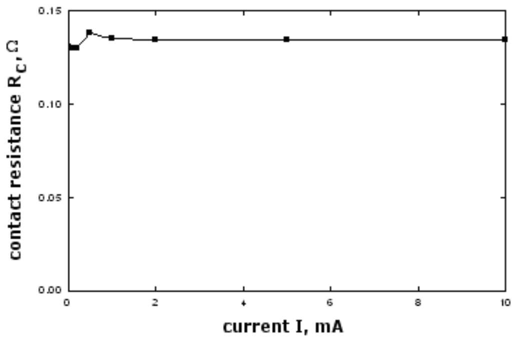

Fig.3. R_c of Ni-Ag interlayer at the liquid nitrogen temperature

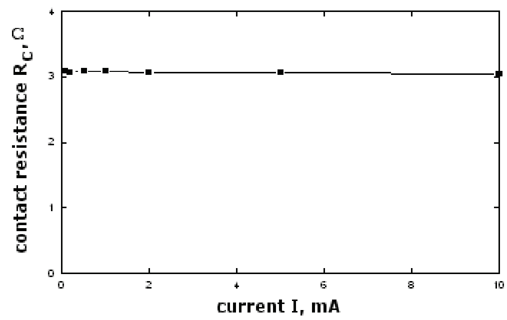

Fig.4. R_c of Ni-Mo interlayer at the room temperature

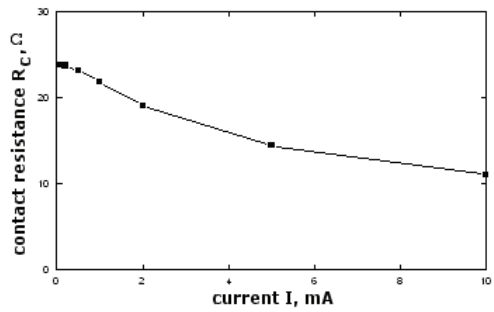

Fig.5. R_c of Ni-Mo contact at the liquid nitrogen temperature

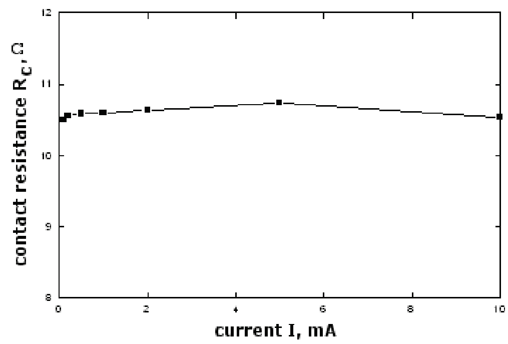

Fig.6. R_c of Ni-Cu interlayer at the room temperature

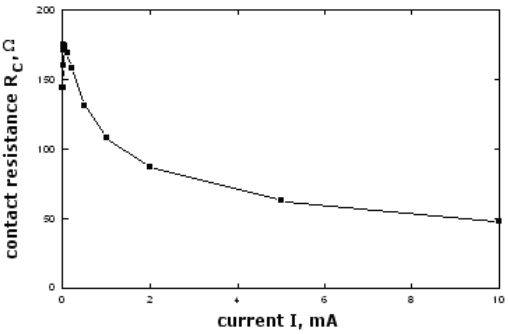

Fig.7. R_c of Ni-Cu interlayer at the liquid nitrogen temperature

For current value I from range of 0-10 mA measured for mono-layer Ni contact the resistance R_c was about 6.5 Ω at the liquid nitrogen temperature (77 K). For higher value of current flow resistance R_c rapidly dropped to the value below 1 Ω. At the room temperature (293 K) R_c value was almost constant (about 1 Ω) during current changes [4].

At the room temperature contact resistance R_c of each double layer is practically constant for the current range 0-10 mA. However, $R_c(I)$ characteristics are quite different at the temperature of liquid nitrogen. As can be seen in Fig. 3, 5 and 7, the values of R_c decrease rather rapidly in each case with increasing of current values. A similar phenomenon was observed in the case of pure Ni layers [4].

It seems that the reason for such behaviour is the high value of resistance R_c at low temperature. The Joule's heat generated in the area of metal/superconductor contact can not be effectively removed by the coolant (liquid nitrogen). This causes a local temperature increase and decrease of metallic contact resistance. The same mechanism may be responsible for additional reduction in value of the measured critical temperature T_c.

In the Table 1 values of contact resistance for mono-layer Ni contact sample [4] and for particular bi-layer samples at two points of their $R_c(I)$ characteristics are collected.

From this overview one can see two features of the tested samples. First, the resistance of the electrode increases with decreasing of the temperature. Second, the resistance decreases with increasing of the value of the flowing current. Both of these characteristics are typical for all samples.

Moreover, not so good adhesion of the layers to the superconductor was found - after several cycles of cooling and heating, they detached from the sample.

Adding another layer to the layer of Ni did not lead to improvement of the contact properties. The only positive result was a slight decrease in resistance in some cases.

Table 1. Contact resistance for particular samples

Contact type	R_c [Ω] for I=1 mA	R_c [Ω] for I=10 mA
Ni (T=77 K)	6.50 [4]	6.50 [4]
Ni (T=293 K)	1.10 [4]	1.00 [4]
Ni-Ag (T=77 K)	0.135	0.130
Ni-Ag (T=293 K)	0.055	0.055
Ni-Mo (T=77 K)	22	11
Ni-Mo (T=293 K)	3.10	3.05
Ni-Cu (T=77 K)	110	50
Ni-Cu (T=293 K)	10.6	10.6

Conclusions

As is known the best electrodes are obtained from silver paint exposed to additional heating. In this paper we have presented the simple electrodes made of mono-layer nickel and of bi-layer Ni-Ag, Ni-Mo, Ni-Cu.

The mono- and bi-layer thin films of Ni, Ni-Ag, Ni-Mo, Ni-Cu were deposited on the surface of $YBa_2Cu_3O_x$ superconductors by magnetron sputtering combined with the evaporation.

A relatively high resistance for metal-superconductor contact characterized each of the obtained layers.

It can be caused not only by electrical properties of electrode material but either chemical reaction in the interlayer between superconductor material and electrode metal.

Any cooling of superconductor material being a substrate for deposited layers was not applied during sputtering process. It could provide better adhesion between deposited metallic film and superconductor surface. However higher temperature could lead to the formation of new non-superconducting (semiconducting) interlayer between them and cause degradation of superconducting properties.

Because of relatively high resistance, the Ni, Ni-Ag, Ni-Mo and Ni-Cu thin films deposited on the surface of superconductor can be used as heaters to control the temperature of the material.

Thus, the only proper material for low-temperature high-current electrode is still silver.

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