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CONDUCTIVITY FEATURES OF NANOMODIFIED HTSC STRUCTURES

M.P. Faradzheva[1], *A.V. Prikhod'ko*[1], *O.I. Kon'kov*[2]

[1] Peter the Great St. Petersburg Polytechnic University, St. Petersburg, Russian Federation;

[2] The Ioffe Institute of the Russian Academy of Sciences, St. Petersburg, Russian Federation

The transport characteristics of nanostructures consistng of microcrystalline $YBa_2Cu_3O_{7-\delta}$ powder and nanopowder of the same composition have been studied in the nanosecond duration interval of the pulse voltages applied to the nanostructure. An increase in the critical temperature T_c of the transition to the superconducting state was established to occur at the 20% nanopowder content. The obtained experimental data was interpreted in terms of the percolation theory. This theory holds that the optimal nanopowder content in the nanomodified material causes a rise of the maximum number of micropowder-nanopowder Josephson's contacts. An increase in the structure porosity when the content falls off from the optimum, leads to a break-down of a percolation cluster and to a decrease in the T_c value; moreover, the temperature range where the transition to the superconducting state takes place, spreads.

Keywords: nanostructure, high-temperature superconductivity, percolation cluster, critical temperature

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ОСОБЕННОСТИ ПРОВОДИМОСТИ НАНОМОДИФИЦИРОВАННЫХ ВТСП-СТРУКТУР

М.П. Фараджева[1], *А.В. Приходько*[1], *О.И. Коньков*[2]

[1] Санкт-Петербургский политехнический университет Петра Великого, Санкт-Петербург, Российская Федерация;

[2] Физико-технический институт им. А.Ф. Иоффе РАН, Санкт-Петербург, Российская Федерация

Проведены исследования транспортных характеристик наноструктур, состоящих из микрокристаллического порошка $YBa_2Cu_3O_{7-\delta}$ и нанопорошка того же состава, в наносекундном интервале длительностей импульсного напряжения, приложенного к наноструктуре. Установлено, что при 20%-м содержании нанопорошка наблюдается повышение критической температуры перехода T_c в сверхпроводящее состояние. Полученные экспериментальные результаты трактуются в рамках теории перколяции, согласно которой оптимальная концентрация нанопорошка в наномодифицированных структурах приводит к возникновению максимального количества джозефсоновских контактов микропорошок – нанопорошок. Увеличение пористости структуры при отклонении от оптимальной концентрации приводит к разрушению перколяционного кластера и уменьшению значения T_c, а также к уширению области температур, в которой происходит переход в сверхпроводящее состояние.

Ключевые слова: наноструктура, высокотемпературная сверхпроводимость, перколяционный кластер, критическая температура

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Introduction

Understanding the mechanism of current flow in materials with high-temperature superconductivity (HTSC) is particularly intriguing because of nonlinear electrical properties manifesting by varying the chemical composition of these materials, the nature of interaction between their grains, and the characteristics of the intergranular space. This includes the "modified" nanomaterials that are close to percolation structures [1–3]. Such physical properties of inhomogeneous percolation structures as electrical conductivity, diffusion, charge carrier mobility and magnetization can be described from the standpoint of percolation theory.

Characteristics of the medium and the parameters of interaction between the regions with identical properties play the key role in percolation theory. Electrical properties of such percolation structures, in particular, conductivity, can be controlled. For example, it was established in [2] that silver-based nanostructures have nonlinear current-voltage characteristics, i.e., a negative differential voltage region appears in the $I–V$ curve with increasing applied voltage. Current flow in a superconductor/manganite composite was studied in [3]. It was found that as the ferromagnetic concentration reached the critical value, the percolation paths were disrupted and a network of superconductor – ferromagnetic – superconductor junctions formed. The presence of the network leads to a sharp increase in the specific resistance of the composite and broadening of the superconducting phase transition (with the ferromagnetic properties preserved). Such compounds are regarded as superconductors with a diffuse phase transition temperature (for brevity, we are going to refer to this region of resistances and temperatures as the $R(T)$ transition from now on). If more than 10% of the magnetic material $La_{0.7}Sr_{0.3}MnO_3$ (LSMO) is added to magnesium diboride (MgB_2), the transport characteristics of such a compound depend on current flow through the LSMO – MgB_2 – LSMO – MgB_2 network, which explains why the $R(T)$ transition broadens by 10 K. It was discovered in [4] by direct-current measurements for the HTSC $YBa_2Cu_3O_{7-\delta}$ ceramics obtained by mixing a microcrystalline powder and a nanopowder of the same composition (with a concentration of more than 20%) that both the absolute value of electrical resistance and the temperature of superconducting transition increased (compared with the corresponding properties of microcrystals).

The goal of this study has been to establish the effect of the composition of HTSC $YBa_2Cu_3O_{7-\delta}$ ceramics obtained by mixing powders with different particle sizes on the critical temperature and the width of the transition to superconductivity for HTSC materials.

In view of our goal, we have considered the characteristics of current flow in such structures depending on the nanopowder content and analyzed the transport characteristics of modified HTSC materials based on microcrystalline $YBa_2Cu_3O_{7-\delta}$ powder and on nanopowder of the same composition exposed to nanosecond-range voltage pulses.

Experimental procedure

The experimental samples consisted of superconducting microcrystalline powder of yttrium barium copper oxide (YBCO), obtained via standard ceramic technology by sintering powders of barium carbonate ($BaCO_3$), yttrium oxide (Y_2O_3) and copper oxide (CuO) in the solid phase, and a superconducting YNCO nanopowder[4, 5], obtained by burning the nitrate organic precursors. Yttrium, barium and copper nitrates were mixed in the corresponding mole fractions and dissolved in water in a ratio of 0.03 : 1.00. Glycerol, added in the amount of 0.5–1.5% of the total mass of the aqueous nitrate solution, acted as a "fuel". The resulting solution was evaporated with continuous stirring to a jelly-like state. This precipitate was ignited and burned to form an amorphous crumbly precursor. Next, the product was subjected to thermal treatment at 350–915°C for 1–20 h at these temperatures to remove unburned organic matter and allow for an appropriate particle size distribution to form. The sizes of nanopowder particles were about 35–70 nm

[5], and of the microcrystals obtained by solid phase sintering 0.5–2.0 μm.

XRD analysis of the synthesized compounds and study of the morphology of the samples, as well as of their phase composition were given earlier in [5].

Samples of the modified structures were obtained by mixing the two powder components in a mass ratio. The components were not subjected to sintering to avoid interdiffusion, undesirable chemical reactions, or compression, with the purpose of excluding the mechanical contribution to the given characteristics. We have considered a model system that was a mixture of powders with artificially created weak Josephson coupling formed at natural intergranular boundaries. The normal resistance of these limits is approximately 3 Ω and does not affect the conclusions of the study.

To assess the transport characteristics of the samples, we investigated the temperature dependences of the resistance $R(T)$ in the region of the superconducting phase transition, with durations of the applied voltage pulses in the nanosecond range, using the method described in [6]. The sample was placed in a cylindrical dielectric cell between two pressure contacts made of glassy carbon and squeezed with these electrodes with a pressure of no more than 1 MPa. Pulse duration was 1–20 ns at voltages up to 1 V at a frequency of 100 Hz. The advantage of this nanosecond experiment is in being able to control the thermal overheating of the current channels, making it possible to detect the current flow patterns in an inhomogeneous system without disrupting it. The resistance of the sample in the measuring cell was adjusted to 100 – 200 Ω (at room temperature) by squeezing it with glassy carbon electrodes; this was necessary to maintain the greatest sensitivity of the technique. The degree of squeezing did not affect the packing density of the powder sample, since the applied forces were negligible (this is confirmed by the fact that we have obtained reproducible results for the same sample). The critical temperature T_c was defined as the point where the transition started. This point for a resistive transition was approximately 94 K for a system with a 100% microcrystalline $YBa_2Cu_3O_{7-\delta}$ powder, which is close to the Curie temperature found via magnetic measurements in [7], and this value is standard.

Experimental results and discussion

Fig. 1 shows the temperature dependences of the sample resistance $R(T)$ in the region of transition to superconductivity for samples with different nanopowder contents (0–100%), with 5 ns

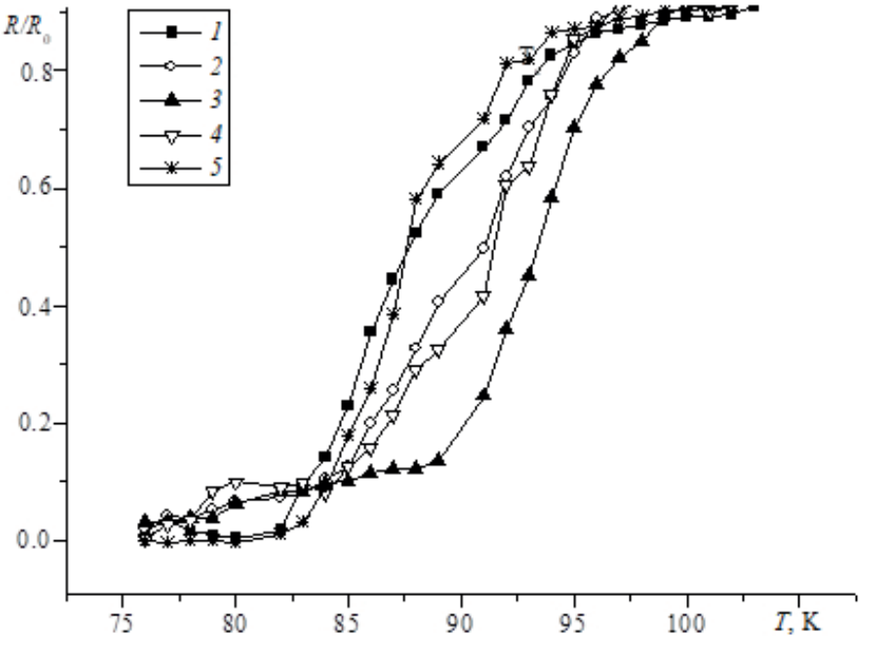

Fig. 1. Temperature dependence of resistance of $YBa_2Cu_3O_{7-\delta}$ samples in the superconducting transition region for different concentrations of the nanopowder added, %: 0 (*1*), 10 (*2*), 20 (*3*), 30 (*4*), 100 (*5*)

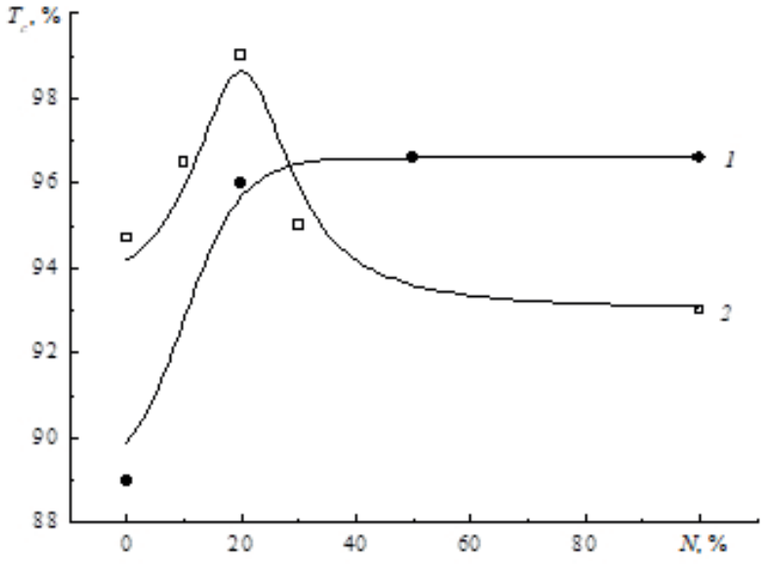

Fig. 2. Critical temperature of superconducting transition as a function of nanopowder concentration in static (*1* [5]) and nanosecond (*2*) modes; the data were obtained for ceramics (*1*) and for a mixture of powders (*2*)

durations of applied voltage pulses. Measurement results are given in relative units, where R_0 is the resistance value at $T = 100$ K.

Studies in static mode [5] showed that the temperature of the transition to superconductivity is 89.0 K for $YBa_2Cu_3O_{7-\delta}$ ceramics obtained by solid phase sintering, and adding nanopowder in a concentration of more than 20% raises this temperature to 96.6 K. However, nanosecond studies revealed an increase by about 5 K in the critical temperature T_c observed only for the sample with a 20% content of nanopowder, while no significant increase in T_c was detected for other concentrations.

Fig. 2 shows the dependences of T_c on nanopowder concentration (N, %) in static (curve *1*, [5]) and nanosecond (curve *2*) modes. The T_c values for purely microcrystalline samples ($N = 0$%) differ in static and nanosecond modes, since the authors of [5] considered $YBa_2Cu_3O_{7-\delta}$ ceramics obtained by solid-phase sintering, and we considered powders in this study.

The critical temperatures (T_c, K), the widths of the superconducting transition (ΔT, K) and the slopes of the temperature dependence of resistance (B, K^{-1}) for samples with different nanopowder contents (N, %) are given in Table. The slope of the *R(T)* curve (parameter *B*) was calculated as the angular coefficient of the tangent at the point of the curve, where the R/R_0 value was half the maximum, i.e., (1/2) $(R/R_0)_{max}$ (the middle of the superconducting transition), with an error of 5%. We found that the slope of the curve for resistance versus temperature for sample *3* with a 20% content of nanopowder differs from other samples (*1*, *2*, *4*, *5*).

The value of B is higher for the sample with a 20% content of nanopowder than for samples of other compositions, which indicates a sharper transition to superconductivity. This composition is also the one for which the highest T_c value, equal to 99 K, is observed.

It was found in [8, 9] that adding $BaTiO_3$ [8] and $BaHfO_3$ [9] nanoparticles to the microcrystalline $YBa_2Cu_3O_{7-\delta}$ compound leads to an increase in the superconducting transition temperature to 103 and 107 K, respectively.

While adding nanoparticles of other elements into YBCO ceramics typically reduces the critical temperature T_c of the transition to superconductivity [10–12], the T_c value increases for our nanostructured materials.

According to percolation theory, conductivity in inhomogeneous media depends on the impurity concentration in the initial sample and on the threshold value of this impurity (N_c is the critical concentration at which the percolation threshold for current is observed). Nanodispersed powder acts as a binding agent in the given structures, forming a network of weak Josephson junctions in microcrystalline HTSC. Nanodispersed filler located between HTSC microparticles allows for a superconducting percolation cluster to form in an inhomogeneous medium.

Table

Parameters of superconducting phase transition for samples of modified $YBa_2Cu_3O_{7-\delta}$ with different nanopowder contents

N,%	0	10	20	30	100
T_c, K	94.7	96.0	99.0	95.0	93.0
ΔT, K	14	14	10	12	13
B, K^{-1}	0.064	0.064	0.090	0.070	0.071

Notations: T_c is the critical temperature; ΔT is the width of the superconducting transition; B is the slope of resistance versus temperature curve; N is the nanopowder content.

The threshold value of the concentration for which an increase in T_c is observed (see Fig. 2) falls on the 20% content of the nanopowder; "switching" of the conductivity mechanism likely occurs in the system above this value. Percolation theory holds that a maximum two-component density (minimum porosity) exists in a bulk two-component system with a 26% volume fraction of smaller particles [13]. In this case, favorable conditions can be generated for Josephson properties to manifest. This concentration leads a maximum number of Josephson junctions between micropowder and nanopowder appearing in nanomodified structures. With increasing concentration, the nanodispersed filler envelops the $YBa_2Cu_3O_{7-\delta}$ microcrystals, forming a shell of a nanopowder with the same composition. A "layer" of nanopowder forms if N is greater than 20%, and direct contact between the micrograins disappears. If the concentration deviates from the optimal, increased porosity of the structure leads to disruption of the Josephson network, fracture of the percolation cluster and to the decrease in the T_c value that we have observed; in addition, the temperature region where the transition to superconductivity occurs is broadened. Notably, this effect is absent in case of constant currents due to prolonged heating.

Conclusion

Studying the transport characteristics for nanosecond voltage pulse durations, we have established the patterns in the evolution of temperature and transition slope of HTSC structures with inhomogeneous composition. We have observed an increase in the critical temperature Tc of the superconducting transition and a narrowing of the superconducting transition at a nanopowder concentration of about 20% both for structures consisting of a microcrystalline $YBa_2Cu_3O_{7-\delta}$ powder and nanopowder of the same composition. Adding nanopowder in various concentrations allows to control the critical temperature and the width of the superconducting transition.

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THE AUTHORS

FARADZHEVA Mislimat P.
Peter the Great St. Petersburg Polytechnic University
29 Politechnicheskaya St., St. Petersburg, 195251, Russian Federation
mpfaradzheva@physics.spbstu.ru

PRIKHOD'KO Aleksander V.
Peter the Great St. Petersburg Polytechnic University
29 Politechnicheskaya St., St. Petersburg, 195251, Russian Federation
aleks@physics.spbstu.ru

KON'KOV Oleg I.
The Ioffe Institute of the Russian Academy of Sciences
26 Polytekhnicheskaya St., St. Petersburg, 194021, Russian Federation
oleg-l705@yandex.ru

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СВЕДЕНИЯ ОБ АВТОРАХ

ФАРАДЖЕВА Мислимат Пиралиевна – *ассистент Института физики, нанотехнологий и телекоммуникаций Санкт-Петербургского политехнического университета Петра Великого.*
195251, Российская Федерация, г. Санкт-Петербург, Политехническая ул., 29
mpfaradzheva@physics.spbstu.ru

ПРИХОДЬКО Александр Владимирович – *доктор физико-математических наук, профессор Института физики, нанотехнологий и телекоммуникаций Санкт-Петербургского политехнического университета Петра Великого.*
195251, Российская Федерация, г. Санкт-Петербург, Политехническая ул., 29
aleks@physics.spbstu.ru

КОНЬКОВ Олег Игоревич – *кандидат физико-математических наук, старший научный сотрудник лаборатории физико-химических свойств полупроводников Физико-технического института им. А.Ф. Иоффе РАН.*
194021, Российская Федерация, г. Санкт-Петербург, Политехническая ул., 26
oleg-l705@yandex.ru