Analysis of technology methods and study of properties of high-temperature superconducting ceramics based on YTTIRIUM

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Abstract. Based on the analysis of the studied properties, the paper presents the developed technology for the manufacture of high-temperature superconducting (HTSC) ceramics based on ultrafine powders; in this case, the method of dry pressing under the influence of an ultrasonic signal is considered. Based on the analysis, the optimal technology variants for synthesizing HTSC ceramics were selected. Data are presented on the performance properties of HTSC ceramics, samples of electromagnetic field screens, volumetric microwave resonators, and ceramic squids. Also, a method of positron annihilation is described, measuring the lifetime of positrons and measuring the curves of the angular distribution of annihilation photons (ADRP), which makes it possible to identify defects of the vacancy type (vacancies, clusters of vacancies, loops, dislocations).

1 Introduction

The development of high-temperature superconducting (HTSC) ceramics is very topical. The international scientific community has reached a temperature slightly higher than the temperature of liquid nitrogen (77 K) for the critical temperature of the superconducting transition Tc. If results above this value are achieved, it will be possible to use HTSC materials in various industries in electronics, electrical engineering, transport, and other areas of the economy.

Therefore, research was developed in the fundamental direction and the applied aspect [1]. The key problem was the development of technologies that make it possible to manufacture the required products from such fragile ceramic materials as complex cuprates: wires and cables, inductors, cavity resonators, etc. In many cases of "low-voltage" applications (electronics, sensors), the use of film technologies for fabricating structures based on HTSC films proved effective. However, for "high-current" applications (energy, transport, accelerator technology, etc.). This article presents the main results of research on the development of methods for manufacturing and studying the properties of HTSC

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ceramics of the $YBa_2Cu_3O_{7+x}$), $(Bi_{0,8}Pb_{0,2})St_2Ca_2Cu_3O_{10}$ and $Bi_{1,7}Pb_{0,3}\,Sr_2CaCu_2O_y$, its compaction and sintering of monophase superconducting ceramics with high critical characteristics. Synthesis of the superconducting orthorhombic phase $YBa_2Cu_3O_{7+x}$ (x<0.4-phase "1-2-3") is a multi-stage process of successive chemical and structural transformations of synthesized components. Among many well-known methods for synthesizing HTSC powders, we have developed the method of solid-phase ceramic synthesis [2].

As is known, solid-phase synthesis is a diffusion process. An analysis of its flow and thermodynamic conditions of synthesis showed that when the synthesis temperature of the 1-2-3 phase is lowered below the well-known 950°C, the probability of the formation of a non-superconducting phase 2-1-1 decreases, and sintering of the formed grains of the 1-2-3 phase is prevented, undesirable at the synthesis stage. Such conditions are achieved when using all the reagents or one of them in the form of UDP. It was shown in [3] that it is sufficient to use only copper in the initial charge in the ultrafine state. In the mixture of composition 1-2-3 with the use of UDP Cu, the synthesis temperature decreases to 920°C and the duration of the formation of the HTSC phase decreases by 12 h, which is associated with an increase in the number of nuclei due to the geometric factor - a larger number and area of contacts between UDP Cu and larger particles Y_2O_3 and $BaCO_3$. The intensification of the kinetics of phase formation is due to an increase in the diffusion coefficient of the substance of submicron copper particles (having the maximum stoichiometric content in the charge) at the boundary of the reagents due to their surface activity, defectiveness, and thermodynamic metastability of the structure, as well as efficient splitting of grains of intermediate phases of synthesis from larger particles of reagents due to intergranular stresses. As a result, a single-phase UDP HTSC $YBa_2Cu_3O_{7+x}$ is synthesized with an average particle size of 0.4-0.7 μm, a critical temperature of the superconducting transition T(c)= 95 K, and a width of this transition Δ Tc = 1 K [2-4].

The formation of the compound $YBa_2Cu_3O_{7+x}$ with such an exothermic effect is possible by the reaction:

$$\frac{1}{2}Y_2O_3 + 2BaO_2 + 3Cu + nO_2 \rightarrow YBa_2Cu_3O_{7+x} + Q,$$

where BaO_2, O_2-oxidizers; - metallic non-oxidized copper reducing agent.

The use of UDP Cu intensifies the synthesis reaction. It increases its thermal effect Q (which determines the self-sustaining of the reaction in the charge) due to the high stored energy of ultrafine particles.

To determine the patterns of SHS of the 1-2-3 system using UDP Cu, we studied the process in an oxygen flow and air, the possibility of controlling the combustion temperature by introducing an additional oxidizer, the degree of compaction of the initial charge, and selecting the sample geometry. In these studies, the task was to determine the conditions under which the combustion temperature lies within 900 ... 970°C, i.e., corresponds to the temperature of synthesis and sintering of the HTSC phase 1-2-3.

2 Objects and methods of research

$YBa_2Cu_3O_{7+x}$ samples were synthesized in different ways and were prepared in the form of pellets 14 mm in diameter and about 3 mm thick. Five series of samples were studied: the first series was pressed under a pressure of $6{,}2N/sm^2$ from a superconducting powder (T_c=91 K) obtained in an oxygen flow by the method of self-propagating high-temperature synthesis (SES) from a $Y_2O_3 - BaO_2 - Cu$ charge. The second series consisted of the same samples, but the conditions for measuring the ADAP curves differed. The survey was

carried out under cryostatting (77...300 K). The third series of samples was prepared using the nitrate technology (T_c=85 K) under a pressing pressure of 3 N/sm^2. The fourth series of samples was a pressing of yttrium-barium cuprite powder (TU48-0531-375-97) at a pressing pressure of 6.4 N/sm^2. The fifth series of samples was irradiated by HEB at the Tonus accelerator at the Tomsk Polytechnic University with the following parameters: energy 1.3 MeV; current density j=1.2 kA/sm^2, pulse duration τ=50 ns. The ADAP curves were measured on a setup with a conditional resolution of 1 mrad and a NaJ positron source with an activity of 7 mCi. Temperature measurements were carried out in an annihilation chamber, in a vacuum in the temperature range (77–300) K, which made it possible to carry out measurements in the region of the critical temperature of the HTSC Ts. Each spectrum was measured for 16÷18 hours, and the total score under the curve was $(1{,}5-6)\cdot 10^6$ counts. The positron lifetime (PLR) was measured on a fast-fast coincidence spectrometer with a time resolution of 225 ps (width at half height). After subtracting the annihilation lifetime in the source substrate from the spectrum $\tau_1 = 150ps, \tau_2 = 450ps$, the spectrum was decomposed into two components. A helium cryostat was used, which allowed changing the temperature from 10 to 300 K. The temperature stability was no worse than 0.5 K. Phase 1-2-3, unreacted $Y_4Ba_3O_9$, $BaCuO_2$, as well as a small amount of orthorhombic phase 1-2-3. The content of the HTSC phase C(sp) increased to 40% after annealing at 950°C for 2 h and to 50–60% after annealing at 950°C for 6 h [1, 5].

For the study, samples were made in the form of tablets with a diameter of 11.2 mm, pressed under pressure, by two methods: statistical mode and when exposed to ultrasound. The USW intensity was set by the output voltage of the US generator U_{USW} 50.75 and 100 V, which corresponded to the amplitudes of the mold wall oscillations A_{USW}=5.10 and 15 μm at a frequency of 21.5 kHz. The samples were baked for 48 hours at different temperatures for different samples: for samples based on UDP-copper at a temperature of: 890°C, and for samples based on standard reagents at a temperature of 950°C. The results of the research can be found in Fig. 1.

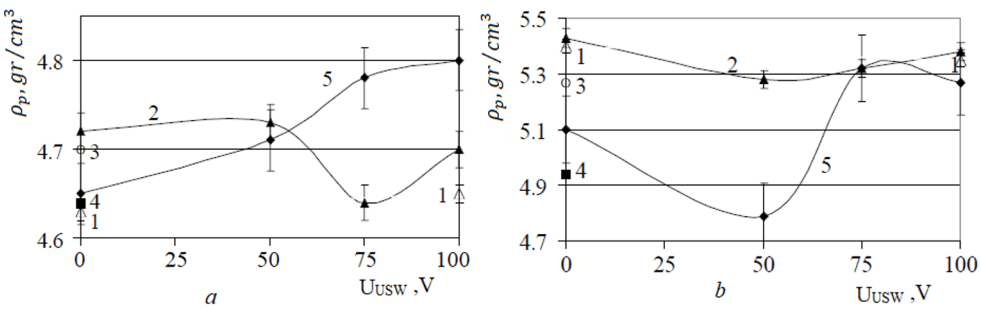

Fig. 1. Density of compacts p- (a) and sintered HTSC ceramics p- (b) depending on intensity of ultrasonic waves and pressing pressure (P) UDP HTSC: 1) 746 MPa; 2) 907 MPa; 3) 1069 MPa; and charging from standard reagents: 4) 746 MPa; 5) 907 MPa

Values of the Meissner effect, which correlates with the content of the HTSC phase in the samples, depend on the charge pressing pressure before SHS initiation.

Our studies have shown that the formation of texture during sintering of 1-2-3 ceramics from the synthesized UDP HTSC proceeds optimally at a pressure of uniaxial dry pressing above 300 MPa, a duration of pressing under such a load of more than 10 h and a sintering temperature of 950 − 975 °C [1, 5].

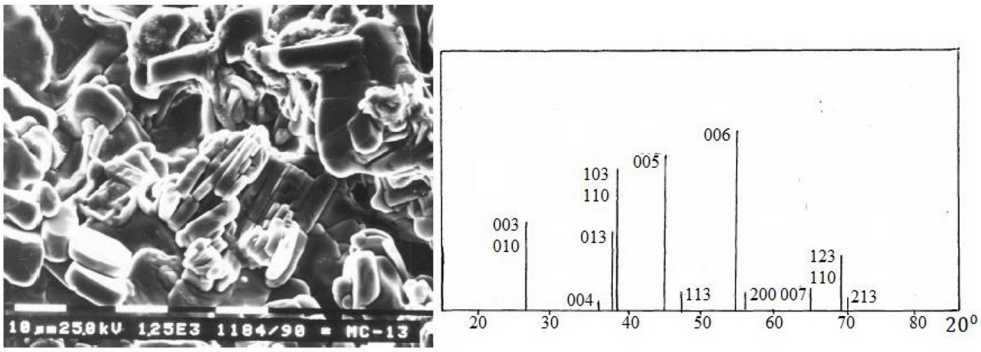

Fig. 2. SEM image of textured HTSC ceramics 1 2 3 sintered from UDP after preloading during pressing and bar graph of X-ray phase analysis (CoK_α radiation)

3 Electrophysical properties of HTSC ceramics and developed products

Testing of superconducting and other physical properties of samples of HTSC ceramics and developed products (HTSC squids, screens of electromagnetic fields, cavity resonators) was carried out by us on calibrated installations by the inductive method (Tc; ΔT_c), 4-pin method (Tc; ΔT_c); critical current j_c), as well as on specialized equipment at the Laboratory of Neutron Physics, JINR (Dubna); at the Laboratory of Microwave Radio Engineering MIREA (Moscow); at the Research Institute of Nuclear Physics at TPU, Research Institute of Semiconductor Devices, Siberian Institute of Physics and Technology at TSU, (Tomsk). Figure 1 presents the results of measurements of the parameters of HTSC ceramic samples fabricated by the technology described above [1, 5].

Table 1. Electrophysical properties of samples of HTSC ceramics

Parameter	ρ_c g/sm^3	$d_з$,* mkm	T_c, K	ΔT_c K	j_c,** A/sm^2	Q_1	Q_2
Ceramics 1-2-3 based on UDP Cu	5.9…6.0	10…20	95	3.5	920	150 250	150 241
Ceramics 1-2-3 from standard reagents	5.2…5.5	40…50	90	1.5	90	–	–

Where *$d_з$, is the average grain size according to optical and scanning electron microscopy data; **j_c is the critical current density determined by the 4-probe method (77 K, 0 T); Q_1 is the quality factor of polished ceramic samples at a frequency $f = 3\,GHz$ ($2\Delta f = 20\,MHz$) at room temperature (in the numerator) and at 77 K (in the denominator), measured at the Laboratory of Microwave Radio Engineering MIREA O.M. Oleinik; Q_2 is the quality factor of the same specimens, measured under the same conditions one year later, indicates the resistance of ceramics to degradation.

Tests of the same cylindrical samples as screens for electromagnetic fields were carried out at the SPTI at TSU.

A technique was used to measure the voltage U_c on the receiving (external) inductance coil located outside the HTSC cylinder when the test current I_t passed through the generating (internal) coil placed inside the hollow HTSC cylinder [1, 5]. Dependencies $U_c = f(I_t)$ were taken in the superconducting state of the screen (T=77 K) and the normal state (at 293 K) Fig. 3.

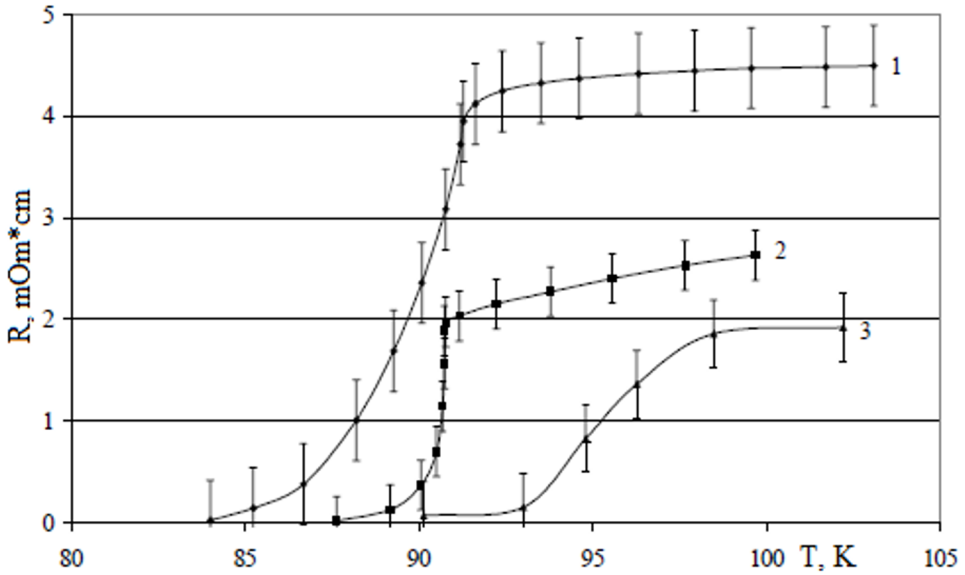

Fig. 3. Curves of superconducting transition for HTSC ceramics fabricated using UDP Cu: 1, 2) dry static pressing, sintering at 920°C and 950°C, respectively (measurements of T_c were carried out at the FLNP JINR by V.N. Polushkin); 3) Ultrasonic pressing, sintering at 950°C (measurements of T_c were carried out at LSHFHR MIREA by A.A. Bush)

The screening factor at T=77 K at a frequency of 10 kHz was k>100. The hysteresis of the field-voltage characteristic (VFC) of the HTSC screen at 77 K, in contrast to the constant at 300 K, also indicates the diamagnetic properties of the product under study (current through the sample I_{cm}=1.3mA; f=10kHz). The sensitivity of a superconducting quantum interference sensor (SQUID) is characterized by the parameter β:

$$\beta = 2 \cdot L \cdot \frac{I_c}{\Phi_o}$$

Here L 10^{-9}…10^{-10} H is the inductance of the quantization circuit in ceramic SQUIDs, which is usually a hole with a diameter of 0.5…1.0 mm; I_c is critical current through the Josephson junction (JC); Φ_o=2.07· 10^{-15} V is magnetic flux quantum. For HTSC squids, the values β= 1…2 are realistically achievable. Therefore, the value of I_c should be 1^{-10} mA. For HTSC ceramics, the values of the critical current density $j_c = \frac{I_c}{s} = 10 \dots 10^3\ {}^{A}/_{sm^2} = 0.1 \dots 10\ {}^{\mu A}/_{\mu m^2}$ at an operating temperature of 78 K (S is the area section of HTSC ceramics). It follows from here that the cross-sectional area of the DP in the SQUID should be within 0.1…100 μм^2,, i.e., the characteristic dimensions of the DP should be 0.3...10 μm [1, 5]. This condition determines the average grain size of HTSC ceramics. To form a DP in HTSC ceramics from grains of the specified sizes, we used the methods of solid-phase synthesis and dry pressing described above to manufacture ceramic HTSC-Squid-Zimmermann type. DP was formed in an HTSC tablet between two holes during the formation and sintering of a dense textured HTSC ceramic with a density of 5.7–6.0 ${}^{gr}/_{sm^3}$ with grain sizes in the texture plane of 10–20 μm. Then, by mechanical scribing under control under an optical microscope and subsequent heat treatment in an oxygen

flow, the required DP thickness of ~10 μm was achieved. The sensitivity of SQUIDs to an external magnetic field reached values of 1...2 μV/Φ_0 [6-10].

4 Conclusions

Under natural conditions, SHS of both a bulk charge of composition 1-2-3 and compacts does not lead to the formation of an HTSC phase, the synthesis of which requires additional annealing at $950^{o}C$.

The initiation of SHS in the air by an electric pulse from the surface of the compacts of the investigated geometry is observed only for a charge with UDP Cu; the use of coarsely dispersed copper, in this case, does not provide the necessary thermal effect of the reaction.

For the formation of an HTSC phase by the SHS method, reagents of a grade no worse than "analytical grade" (primarily, the BaO_2) oxidizer) are required.

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