

PERCOLATION EFFECT IN THICK FILM SUPERCONDUCTORS

(Using a Bi(Pb)SrCaCuO based paste to prepare a superconducting planar transformer)

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SUMMARY

A thick film superconductor paste has been developed to study the properties of granulated superconductor materials, to observe the percolation effect and to confirm the theory of the conducting mechanism in the superconducting thick films. This paste was also applied to make a superconducting planar transformer. Due to high T_C and advantageous current density properties the base of the paste was chosen to be of Bi(Pb)SrCaCuO system. For contacts a conventional Ag/Pt paste was used.

The critical temperature of the samples were between 110 K and 115 K depending on the printed layer thickness. The critical current density -at the boiling temperature of the liquid He- was between 200 - 300 A/cm². The R(T) and V(I) functions were measured with different parameters. The results of the measurements have confirmed the theory of conducting mechanism in the material. The percolation structure model has been built and described.

As an application, a superconducting planar thick film transformer was planned and produced. Ten windings of the transformer were printed on one side of the alumina substrate and one winding was printed on the other side. The coupling between the two sides was possible through the substrate. The samples did not need special drying and firing parameters. After the preparation, the properties of the transformer were measured. The efficiency and the losses were determined. Finally, some fundamental advantages and problems of the process were discussed.

1. INTRODUCTION

In recent years, various methods were applied to prepare superconductor materials. The screen - printing film technology is one of the rarely applied methods, however it can be performed using conventional substrates and it needs only cheap polycrystalline materials. In Hungary a good manufacturing process was developed to make a high T_C superconductor paste, which does not need special drying and firing parameters. Using the paste, the process is fully compatible with the traditional thick film technology. Thus, the superconductor electronic elements made with this technology can be connected to other thick-film electronic elements and two dimensional layouts of complicated shape can be prepared.[1]

2. STRUCTURE OF THE MATERIAL

The base of the paste is the Bi(Pb)SrCaCuO system. This type of superconductor material has a granulated structure. In this structure, the typical grain size is between 0.5 to 10 μm . The couplings between the individual grains generate a 3 dimensional - net, resulting in superconduction. The name of these couplings, because of their superconductor-insulator-superconductor structure, is "Josephson transitions" and the name of the net is "Josephson - net". Mészáros et al [2] have observed, that the composition of the surface of the grains is different from the inside of the grains. The surface area is richer in copper and poorer in oxygen than the bulk. This difference may cause the relatively low critical current density of the samples.

3. SAMPLE PREPARATION

The samples were prepared on 96 % alumina substrates by the method described by Besenyei et al [3]. During the preparation of the paste, the grain size was measured on the basic powder. Firstly, more than 30 percent of the sample consisted of grain-size 10 - 50 μm . This average size was still not satisfactory for the conducting mechanism, so a manual grinding was applied to decrease the size of grains. Secondly, the grain size distribution was better then in the first case because the average size was about 5 μm .

After the grain size measurements, a printable paste was produced from the basic powder. Samples were printed with 200 mesh screen (the swap off was 1.8 mm) and dried for 10 minutes at 150 °C in air. The superconductor layer was fired in a Johnson -Watkins furnace with a normal temperature profile, where T_{max} was 850°C. For contacts, a general purpose Ag/Pt conductor paste was applied. In the case of contact layers the maximum firing temperature was 800°C and the atmosphere was air, too. The thickness of fired layers were 20 and 38 μm .

4. RESULTS AND DISCUSSION

The electric field in the superconductors depends on the current density (J), on the external magnetic field (B_{ext}) and on the temperature (T). However, the value of the magnetic field (B) is determined not only by the external magnetic field, but also by the so called "magnetic prehistory" of the samples. Before the application of the paste, we wanted to know the electrical properties of the superconductor samples. So, we performed the following measurements by using contacts as shows in Fig.1:

1. R - T function $V=V(T)$, $B_{\text{ext}}=\text{const}$, $I=\text{const}$
2. I - V function $V=V(I)$, $B_{\text{ext}}=\text{const}$, $T=\text{const}$
3. B - V function $V=V(B)$, $I=\text{const}$, $T=\text{const}$

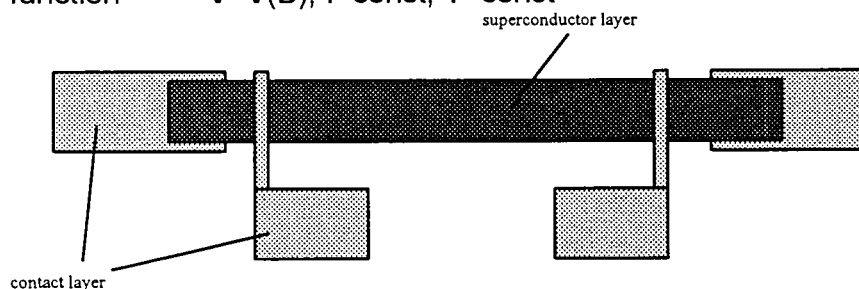


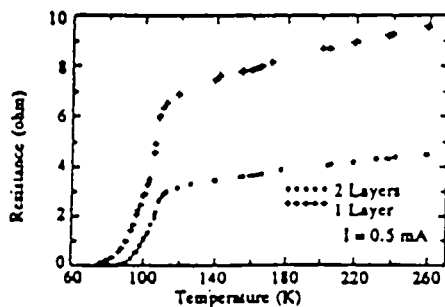
Fig.1 Layout of four-contact method

4.1. The R - T functions

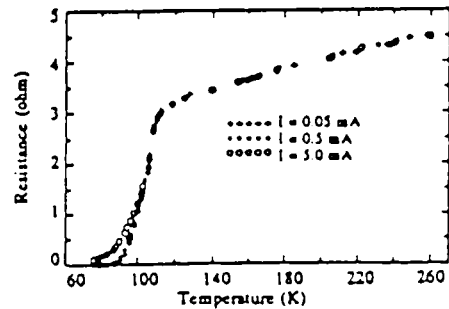
These functions are important characteristic of the superconducting materials, because the T_C can read from the graphs and the graphs show the different ways of the conducting mechanism. Fig.2.a shows two graphs : One corresponds to the one-layer sample, the other to the two-layer sample. One can observe, that the resistance of the two-layer sample is less than the one-layer, but the T_C -s are equal. Thus, the T_C which can be read from the graphs is about 115 K.

Fig.2.b shows the results of measurements with three different current values from 0.05 mA to 0.5 mA by decades. The graphs confirmed the theory of the conducting mechanism. They show a significant difference only in the case of $T < T_C$. The reason may be the grainy structure of the material. The graphs can be divided into two parts according to the way of conducting mechanism;

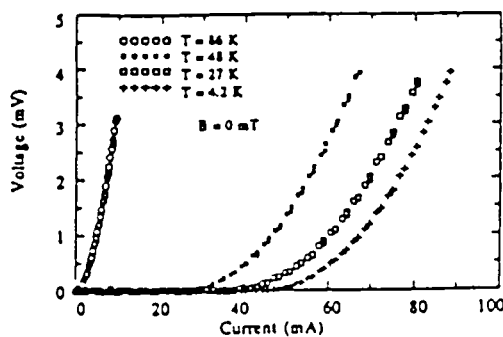
- in the case of $T > T_C$, almost only the individual grains conduct and their critical current density is larger than the critical current density of the inter - grain Josephson - transitions. Thus, the three graphs coincide;
- in the case of $T < T_C$, the inter - grain Josephson - transitions determine the critical current of the samples. This current is less than the critical current of the grains, so a difference between the measuring currents appears on the graphs.



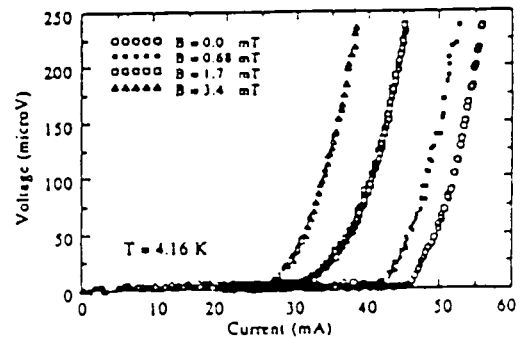
a.



b.



c.



d.

Fig.2 Measurement results by using four-contact method:
a-b. R-T functions; c-d. V-I functions;

4.2. The V- I functions

The graphs shown in Fig.2.c are the V - I functions with T as a parameter, when T ranges from 4.2 K to 86 K and the value of external magnetic field is zero. Significant difference was registered only at T=86 K. Here, the critical current density decreased drastically and the sample resistance increased rapidly to a relatively high level. The reason of this deviation is the low critical current of inter-grain Josephson-transitions.

The V - I function with B as a parameter was measured at constant T=4.2 K temperature. Fig.2.d clearly shows that the external magnetic field works against the superconduction. The value of the critical current decreased significantly with increased of the value of the external magnetic field.

From the results, the so-called "percolation model" can be built up. It means: the 3D-bar ceramic superconductor consists of many individual grains. These grains joint to each other with Josephson-transitions and the grains with the transitions form a weak-coupled net. This 3D-net is the percolation structure. Fig.3 shows a piece of superconductor material in 2D, where the grains and the percolation paths can be observed. During measurements, the most important observation was that the external magnetic field penetrates into the grains -decreasing the critical current density of the samples before its value reaches the value of the critical magnetic field of the superconductor (H_C). The reason of this phenomena may be that a so-called "Josephson-flux" starts to grow between the grains.

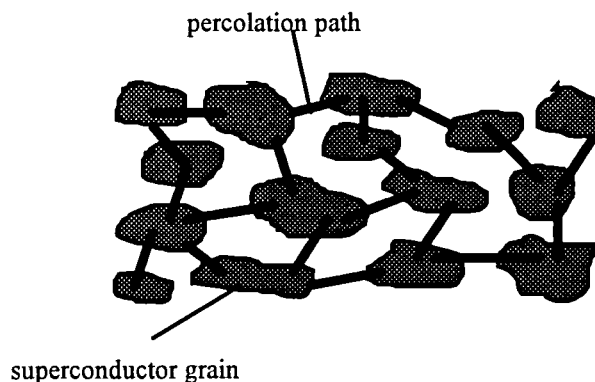


Fig.3 Percolation paths in the superconductor

5. THE PLANAR TRANSFORMER

Finally, a realised superconductor planar transformer is described. The properties of the superconductor layers of the transformer were the same as we stated above. Our aims were as follows;

- to transform the voltage from a higher level to a lower level, where the response is true to the original form;
- to realise a good electrical isolation between the two sides of the transformer.

According to the stated rules above, ten windings of the transformer were printed on one side of the 96 % purity alumina substrate and one winding was printed on the other side as shown in Fig.4. Transformer parameters were as follows [4]:

| | |
|----------------------------------|-------|
| -Internal diameter | 10 mm |
| -Outside diameter | 40 mm |
| -Width of primary turns | 1 mm |
| -Width of secondary winding turn | 15 mm |

The samples were prepared with the same drying, firing and printing parameters as we have described previously, but the primary and the secondary superconductor layers of the transformer were fired at the same time. It was a big problem to give the same atmosphere and the same temperature profile to both layers on two sides. Number of the printed layers were 1 or 2 on each side of the substrates. On the secondary side of the transformer the width of the winding was different from the width of the primary windings. The reason of this layout design was that we wanted a good coupling between the two sides. It was very interesting, that the fully non-magnetic substrate can cause a satisfactory coupling.

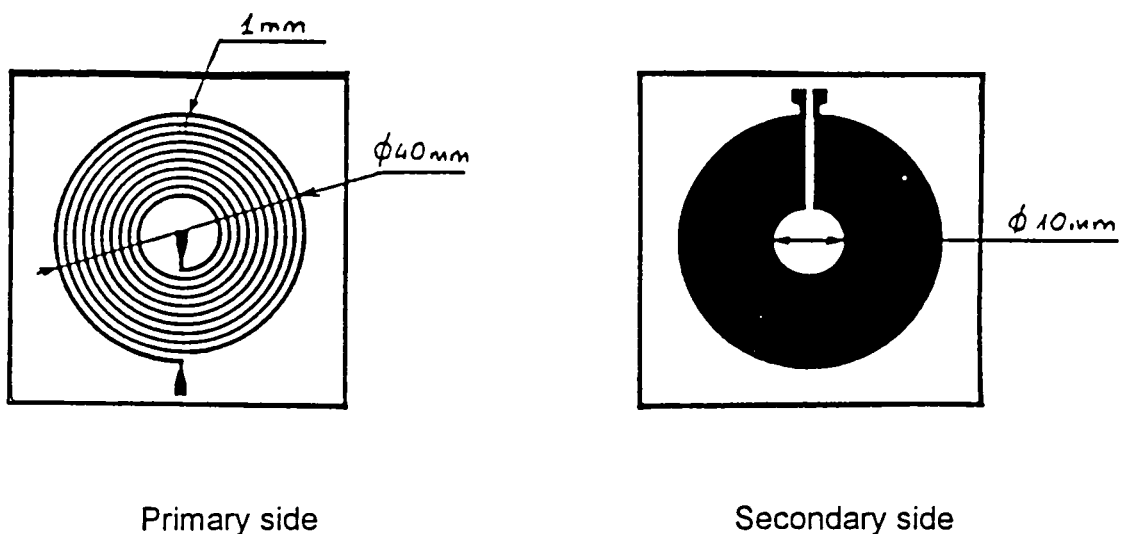


Fig.4 Layout of the planar transformer

5.1. Measurements on the transformer

In the first part of the measurements we wanted to make certain of the superconduction on each side of the transformer. Thus, we performed the simple four - contact measuring on both sides of the samples. The results were very interesting, because we could find the abrupt resistance - changing, but the value of the resistance did not reach zero. There are two possible different explanation of this phenomena;

- because of the two-side firing technology, the samples are not able to reach the superconducting state and they have a finite resistance;

- the samples would be able to reach the superconducting state, if the transformer had only one side. We suppose, that the throw-out magnetic field of the non-measured side of the sample (which is in superconducting state too) may destroy the superconduction of the measured side. This influence prevents the samples from reaching the superconducting state.

In the second part of the measurements the superconductor transformer was measured in normal and in cooled (superconducting) state. At last the measurement results of the superconductor samples were compared with another transformer which was made of a general purpose Ag/Pt conducting paste. The measurements were performed with three different input signals (square-, triangle- and sine functions). We gave to the samples 2.2 V peak-to-peak voltage and examined the output voltage with oscilloscope. The efficiency (η) and the losses (δ) were defined:

$$\eta = \frac{V_{outreal}}{V_{outideal}} \quad (1.) \quad \delta = 1 - \eta \quad (2.)$$

where $U_{outreal}$ was the value of the measured voltage on the secondary side and $U_{outideal}$ was the value of the input voltage divided by the coupling factor ($n=10$). Thus, the efficiency and the losses could be calculated based on the results. The frequency of the input signals was chosen as a parameter from 10 kHz to 2 MHz.

Table 1. shows the measurement results of a two-layer superconductor sample in normal state. We observed, that the transmission of the square- and the triangle functions was not satisfactory because these functions were differentiated by the sample. The transmission of the sine function was faithful to the original form in all frequency - region. Thus, later the sine input signal was chosen to be the main input signal. The maximum efficiency was calculated at 1.9 and 2 MHz, its value was 79%.

Table 1. Measurement results in non-superconducting state

| Frequency (kHz) | Response of 2-layer sample (mV) | Efficiency (%) | Losses (%) |
|-----------------|---------------------------------|----------------|------------|
| 10 | 8 | 4 | 96 |
| 100 | 51 | 23 | 77 |
| 1000 | 130 | 59 | 41 |
| 1300 | 145 | 66 | 34 |
| 1600 | 165 | 75 | 25 |
| 1900 | 175 | 79 | 21 |
| 2000 | 175 | 79 | 21 |

We wanted to measure the one-layer samples too, but after the firing these samples became useless, because the relative thin superconductor layer was not continuous.

Table 2. shows the measurement results of the previous two-layer superconductor transformer in the superconducting state. The main input signal was the sine function with different frequencies from 10 kHz to 2 MHz. One can observe, that the efficiency was better - above 1 MHz - than we measured in the normal state. The maximum efficiency was calculated at 1.9 and 2 MHz, its value was 89 %. The increased efficiency value might be caused by the abrupt resistance decreasing.

Table 3. shows the measurement results of a two-layer planar transformer made of general used Ag/Pt paste. This device had got a very low efficiency and high losses. It started to transform only at 1 MHz, but it had got a big advantage: It worked with 20 V peak-to-peak input voltage too.

Table 2. Measurement results in superconducting state

| Frequency (kHz) | Response of 2-layer sample (mV) | Efficiency (%) | Losses (%) |
|-----------------|---------------------------------|----------------|------------|
| 10 | 6 | 3 | 97 |
| 60 | 30 | 14 | 86 |
| 100 | 48 | 22 | 78 |
| 600 | 120 | 55 | 45 |
| 1000 | 150 | 68 | 32 |
| 1300 | 170 | 77 | 23 |
| 1600 | 180 | 82 | 18 |
| 1900 | 195 | 89 | 11 |
| 2000 | 195 | 89 | 11 |

Table 3. Measurement results of a traditional thick-film transformer

| Frequency (kHz) | Response of the sample (mV) | Efficiency (%) | Losses (%) |
|-----------------|-----------------------------|----------------|------------|
| 100 | 0 | 0 | 100 |
| 600 | 0 | 0 | 100 |
| 1000 | 60 | 27 | 73 |
| 1300 | 80 | 36 | 64 |
| 1600 | 80 | 36 | 64 |
| 1900 | 90 | 41 | 59 |
| 2000 | 100 | 45 | 59 |

6. CONCLUSIONS

The produced paste correctly showed the special properties of the high T_c superconducting ceramics. This paste could be printed well and easily, it did not need special drying and firing methods and parameters. The measurement results of the R-T functions and the percolation effect confirmed the theory of the conducting mechanism in the superconducting thick films.

A superconductor middle- and high- frequency planar transformer was fabricated and tested. This transformer operated satisfactory in the MHz region. It was compared to a normal planar transformer and the results were given. The greatest problem of the transformer was caused by the grainy structure of the superconductor material. In this type of superconductor the boards of grains as a Josephson - transitions are not able to put up higher power. Thus, the conduction mechanism gives a limit of using and the value of the input signal of the transformer was relatively low, 2.2 V. The maximum critical current density was 10^3 A/cm² as opposed to the 10^6 A/cm² ideal value.

The way of improvement is to decrease the size of the grains in order to increase the value of the critical current density.

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