
ID #491

**ICMC 05: Thin films, multilayers, electronics, materials, processing and properties**

# Oxygenation under high pressure of EuBCO and GdBCO coated conductors

**T. Prikhna[1,2,3*], T. Puig[2], A. Kethamkuzhi[2], R. Vlad[2], R. Kluge[3], M. Karpets[1,4], S. Ponomarov[5], V. Moshchil[1], , S. Wurmehl[3], and X. Obradors[2]**

[1] V. Bakul Institute for Superhard Materials of the National Academy of Sciences of Ukraine,
1 2, Avtozavodska Str., Kyiv 07074, Ukraine
[2] Institut de Ciencia de Materials de Barcelona, CSIC, Campus UAB, 08193 Bellaterra, Spain
[3] Leibniz-Institut für Festkörper- und Werkstoffforschung Dresden e. V.,
Helmholtzstrasse 20 01069 Dresden, Germany.
[4] National Technical University of Ukraine «Igor Sikorsky Kyiv Polytechnic Institute”,
Peremogy Avenue 37, 03056 Kyiv, Ukraine
[5] V.E. Lashkaryov Institute of Semiconductor Physics of the National Academy of Sciences of Ukraine,
41, Nauky Ave., Kyiv 03028, Ukraine

# Content

1. **Motivation**

2. **Experimental ( 1-160 bar oxygen pressure, 300-800 $^{o}$C temperature, 3- 12 h holding time)**

3. **Characteristics before and after treatment ($T_c$, $J_c$, carrier density, $n_H$, *c*-lattice parameter of RE123 (RE=Gd, Eu)**

4. **Study of the materials structures (SEM EDS and Auger spectroscopy).**

5. **Conclusions**

# 1. Motivation

- The overdoping for $YBa_2Cu_3O_{7-\delta}$ (Y123) thin films (as a result of treatment via Ag layer when heated under oxygen partial pressure below 1 bar at low temperatures) was confirmed by a sudden increase of charge carrier density, $n_H$, associated to the reconstruction of the Fermi-surface at the Quantum Critical Point (QCP). The authors (Stangl, A., Palau, A., Deutscher, G., Obradors, X., 2021, . Sci Rep., 11, 8176.) achieved the highest ever reported in YBCO films critical current density $J_c$, equal to 90 MA·cm$^{-2}$ at 5 K which corresponds to about a fifths of the despairing current.

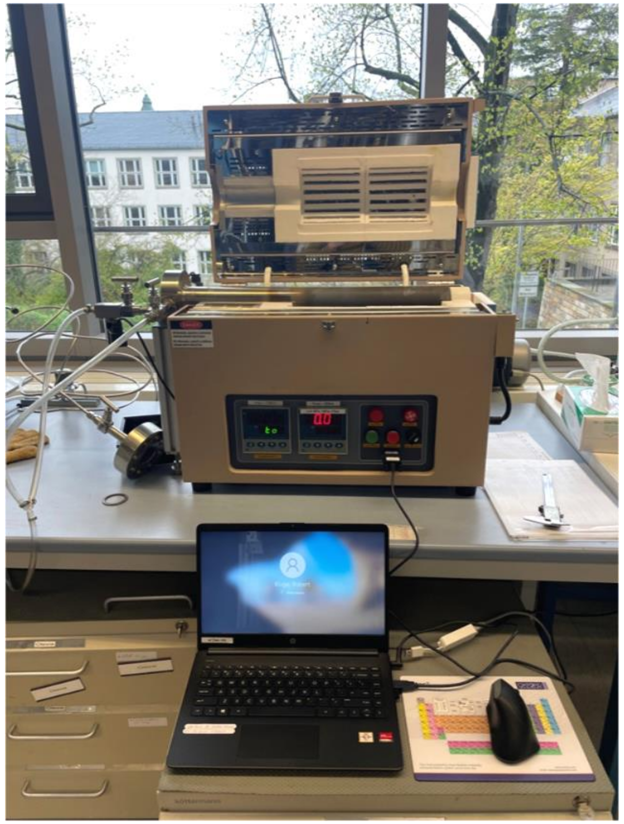
- The Miura, M. et al. ( 2022, NPG Asia Materials , 14, 85) demonstrated that thermodynamic improvements of superconductors can work in parallel with already successful artificial pinning centers and that a maximum critical current density $J_c \sim 0.3\ J_d$ (despairing current ) appears to be the current upper limit for the enhancement in $J_c$. They concluded that variation in $\lambda$ and $\xi$ leads to an intrinsic improvement in $J_c$ via $J_d$, allowing extremely high values of $J_c$ of 130 MA/cm$^2$ at 4.2 K, consistent with an enhancement in $J_d$ of a factor of 2 for nanoparticle-doped (Y,Gd)123 coated conductors (CCs). The combination of thermodynamic and pinning optimization route for the (Y,Gd)123 CCs resulted in high vortex-pinning force ~3.17 TN/m$^3$ at 4.2 K and 18 T (H||c) .

- The idea to apply such high oxygen pressure (100-160 bar) and annealing temperature (600-800 $^o$C) was caused by our experience with melt-texturing YBCO for which such conditions allowed essentially decrease the time of oxygenation, increase critical current density (especially in high magnetic fields) due to increase of twin density, reduce anisotropy of critical current density, decrease mico- and macrocracking (and thus improve mechanical properties). [3-5]

## 2. Experimental

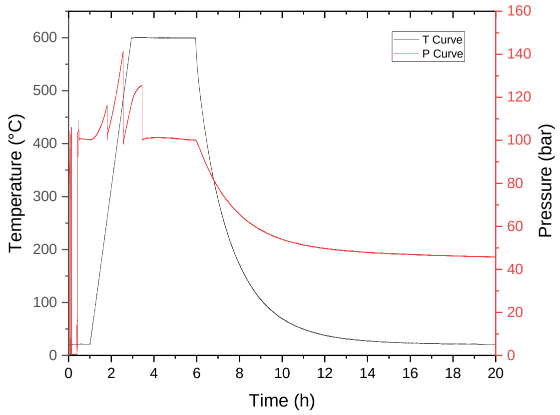
### Architecture of the studied types of coated conductors:

(1) FYSC : Ag (2μm)/ GdBCO (1.8μm)/ $Al_2O_3/Y_2O_3$/MgO/ $CeO_2$ (700 nm)/ Hastelloy (75 μm);
(2) FESC : EuBCO (2.5μm)+BHO Nanorods/ $Al_2O_3/Y_2O_3$/MgO/ $CeO_2$ (700 nm)/ Hastelloy (50 μm);
(3) FESC : Ag (2μm)/ EuBCO (2.5μm)+BHO Nanorods/ $Al_2O_3/Y_2O_3$/MgO/ $CeO_2$ (700 nm)/ Hastelloy 50 μm).

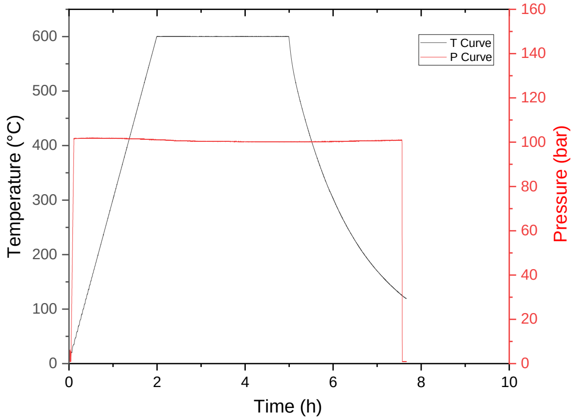


**Tube furnace. 1-160 bar $O_2$, 300-800 $^oC$, $v_{heating}$ = 5K/min, $\tau$= 3-12 h**

**Mode 1**




**Mode 2**

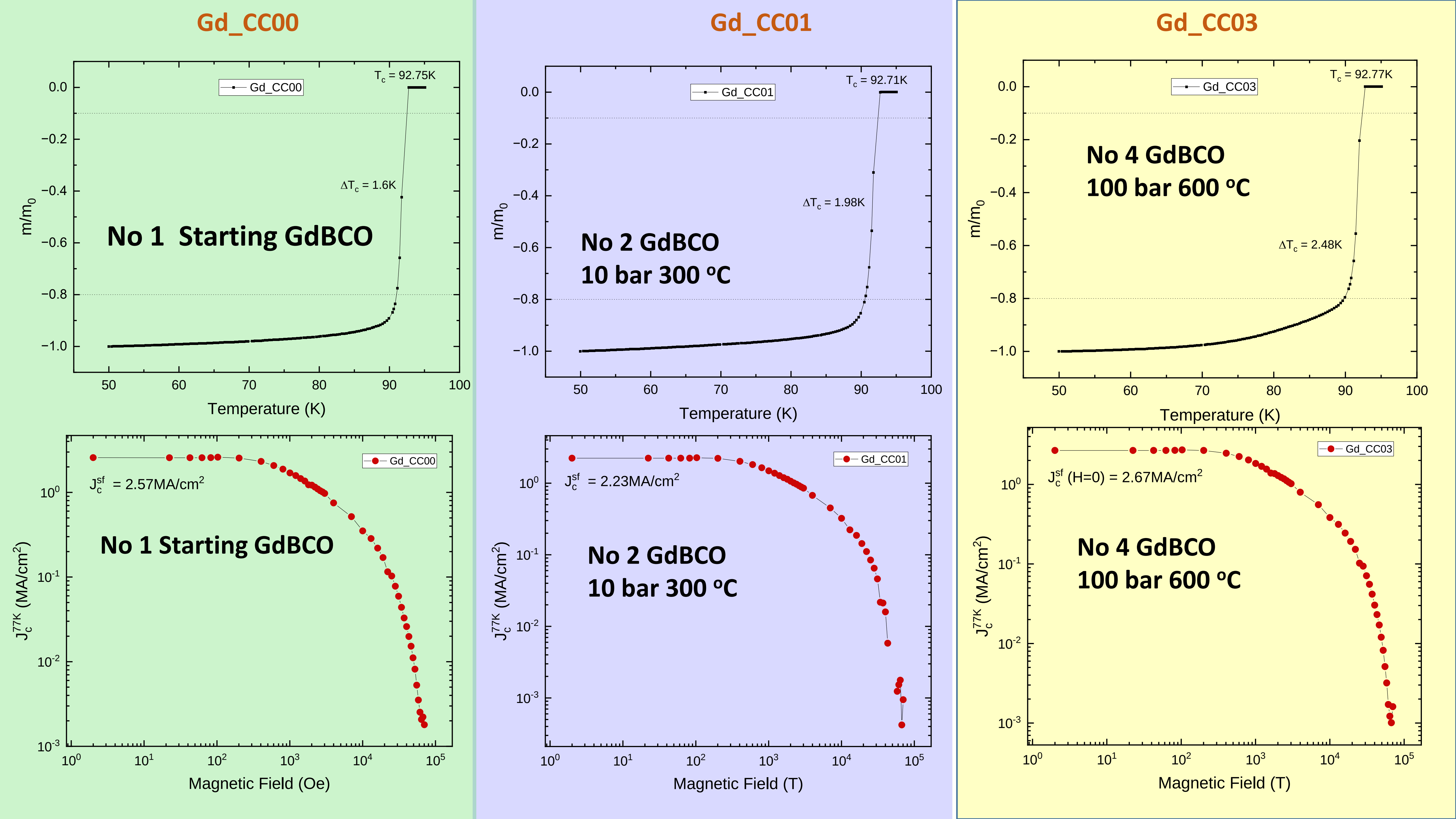



**The transition temperature, $T_C$, and critical current density, $J_c$, were estimated by SQUID magnetometer and $T_c$ for EuBCO-BHO without Ag layer were estimated using transport measurements; charge carrier density, $n_H$, were found from the Hall effect at 100 K.**

**JAMP–9500F, JEOL, Japan –**
**Scanning Auger Microscopy which is combination of HRSEM (secondary electron mode, resolution 3 nm) and Local Auger-electron Spectroscopy (resolution 5 - 8 nm)**

## 3. Characterization

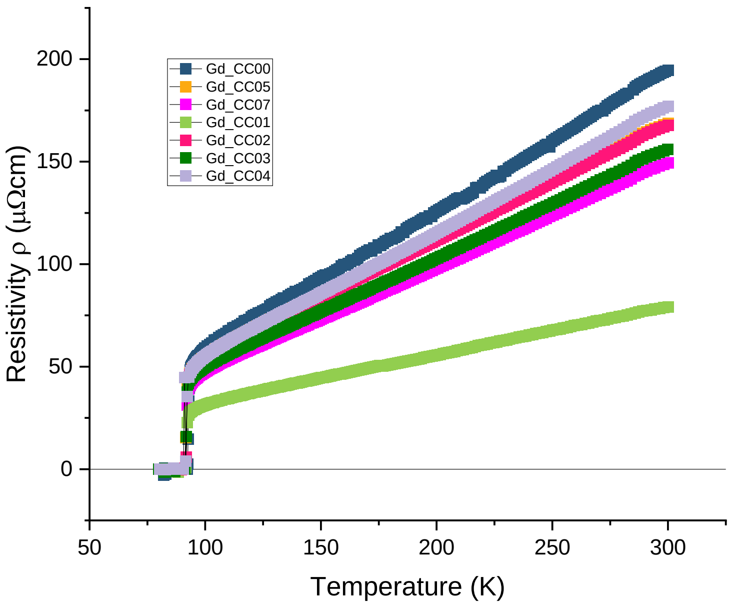
| No | $p(O_2)$, bar | $T_s$, °C | τ, h | Mode 1 or 2 | $J_c$ (0 T, 77K), $MA/cm^2$ | *c*-parameter, nm | $n_H$(100 K), $\times 10^{21}$ $cm^{-3}$ | $T_c$, K |
|---|---|---|---|---|---|---|---|---|
| (1) Ag (2μm)/ GdBCO (1.8μm)/ $Al_2O_3/Y_2O_3$/MgO/ $CeO_2$ (700 nm)/ Hastelloy (75 μm) | | | | | | | | |
| 1 | Starting | | | | 2.57 | 1.17351 | 9.57 | 92.75 |
| 2 | 10 | 300 | 3 | 1 | 2.23 | 1.17318 | 20.76 | 92.71 |
| 3 | 5 | 300 | 3 | 1 | 2.49 | 1.17343 | 11.91 | 92.60 |
| 4 | 100 | 600 | 3 | 1 | 2.67 | 1.17310 | 15.23 | 92.77 |
| 5 | 160 | 800 | 3 | 1 | 2.49 | 1.1,7300 | 12.04 | 92.74 |
| 6 | 2 | 300 | 3 | 1 | 2.47 | 1.17289 | 11.12 | 93.26 |
| 7 | 1 | 300 | 3 | 1 | 2.19 | 1.17320 | 21.90 | 92.76 |
| 8 | 100 | 600 | 6 | 1 | 2.36 | - | 7.64 | - |
| 9 | 100 | 600 | 12 | 1 | 2.65 | - | 43.49 | - |
| (2) EuBCO (2.5μm)+BHO Nanorods/ $Al_2O_3/Y_2O_3$/MgO/ $CeO_2$ (700 nm) | | | | | | | | |
| 10 | Starting | | | | 1.38 | 1.174024 | 6.05 | 92.35 |
| 11 | 10 | 300 | 3 | 1 | 0.87 | 1.174638 | 3.77 | 91.65 |
| 12 | 5 | 300 | 3 | 1 | 0.99 | 1.174309 | 3.85 | - |
| 13 | 100 | 600 | 3 | 1 | 0.77 | 1.174261 | 4.02 | - |
| 14 | 160 | 800 | 3 | 1 | 1.31 | 1.173633 | - | - |
| 15 | 2 | 300 | 3 | 1 | 1.09 | 1.174117 | 4.43 | - |
| 16 | 1 | 300 | 3 | 1 | 1.09 | 1.174015 | 4.33 | 91.65 |
| 17 | 100 | 600 | 3 | 1 | 0.97 | 1.17405 | 4.92 | 92.22 |
| (3) Ag (2μm)/ EuBCO (2.5μm)+BHO Nanorods/ $Al_2O_3/Y_2O_3$/MgO/ $CeO_2$ (700 nm)/ Hastelloy (50 μm) | | | | | | | | |
| 18 | 160 | 600 | 3 | 2 | 1.32 | 1.17488 | - | 93.04 |
| 19 | 160 | 800 | 3 | 2 | 0.69 | 1.17471 | 6.69 | 92.09 |

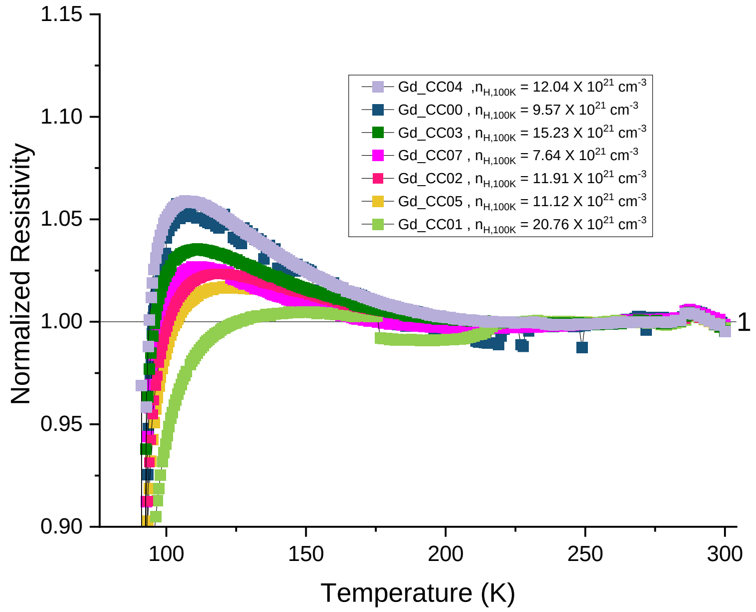


Gd_CC00
Gd_CC01
Gd_CC03
Gd_CC00
T_c = 92.75K
ΔT_c = 1.6K
No 1 Starting GdBCO
m/m_0
Temperature (K)
Gd_CC01
T_c = 92.71K
ΔT_c = 1.98K
No 2 GdBCO
10 bar 300 °C
m/m_0
Temperature (K)
Gd_CC03
T_c = 92.77K
No 4 GdBCO
100 bar 600 °C
ΔT_c = 2.48K
m/m_0
Temperature (K)
Gd_CC00
J_c^sf = 2.57MA/cm^2
No 1 Starting GdBCO
J_c^77K (MA/cm^2)
Magnetic Field (Oe)
Gd_CC01
J_c^sf = 2.23MA/cm^2
No 2 GdBCO
10 bar 300 °C
J_c^77K (MA/cm^2)
Magnetic Field (T)
Gd_CC03
J_c^sf (H=0) = 2.67MA/cm^2
No 4 GdBCO
100 bar 600 °C
J_c^77K (MA/cm^2)
Magnetic Field (T)

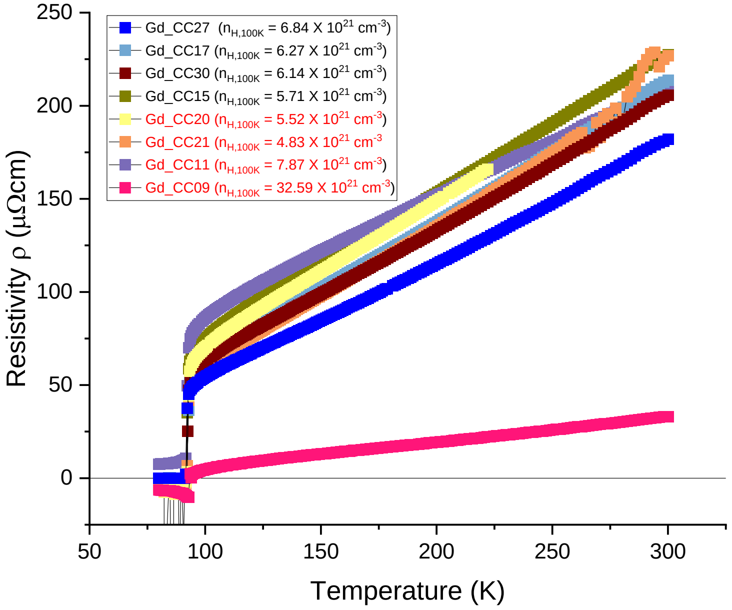
# Comparison of normalized resistivity

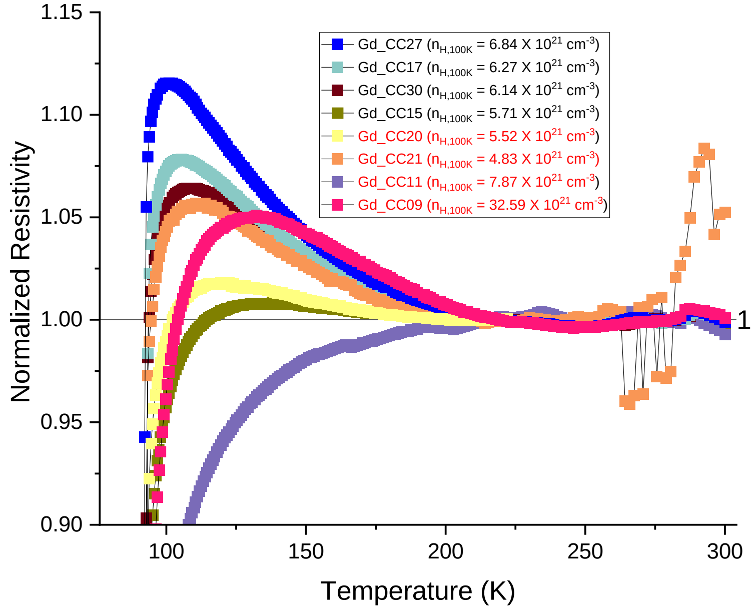
**Mode 1**




**Mode 2**



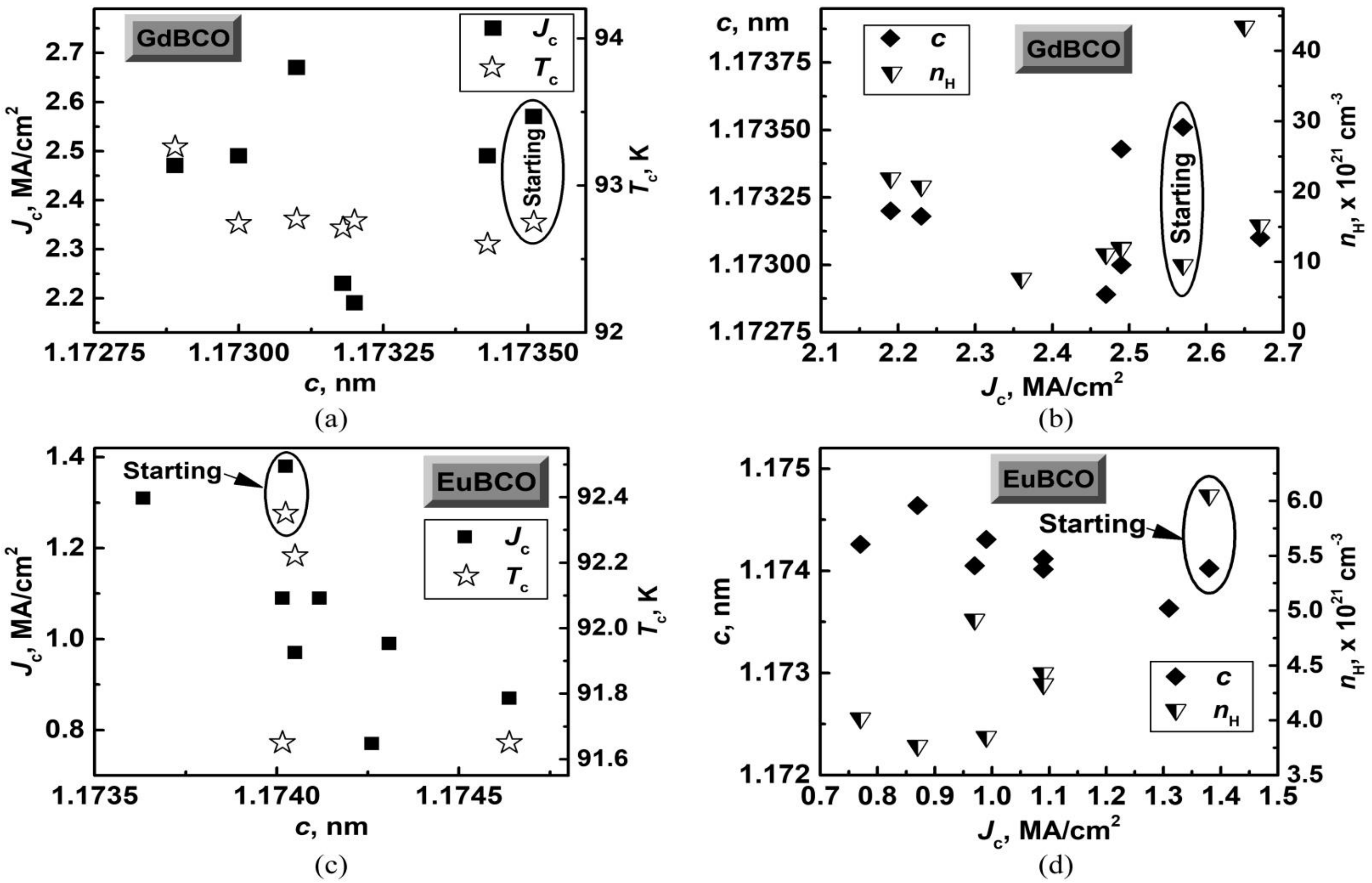
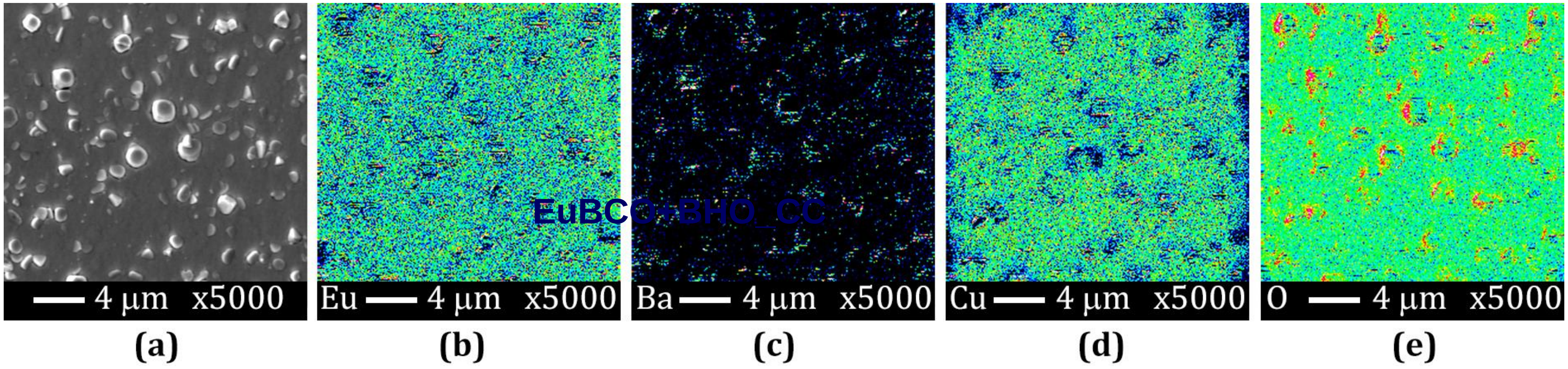



**Figure.** (a, b) – Critical current density, *J*c, at 77K in 0 T and transition temperature $T_c$ vs c-lattice parameters of GdBCO with Ag layer on the top and of EuBCO+BHO nanorods without Ag layer on the top, respectively; (c, d) -*c* -lattice parameters and charge carrier density, $n_H$ (100K) vs critical current density, *J*c, at 77K in 0 T of of GdBCO with Ag layer on the top and of EuBCO+BHO nanorods without Ag layer on the top, respectively.

## GdBCO_CC (starting)




## EuBCO+BHO_CC (starting)

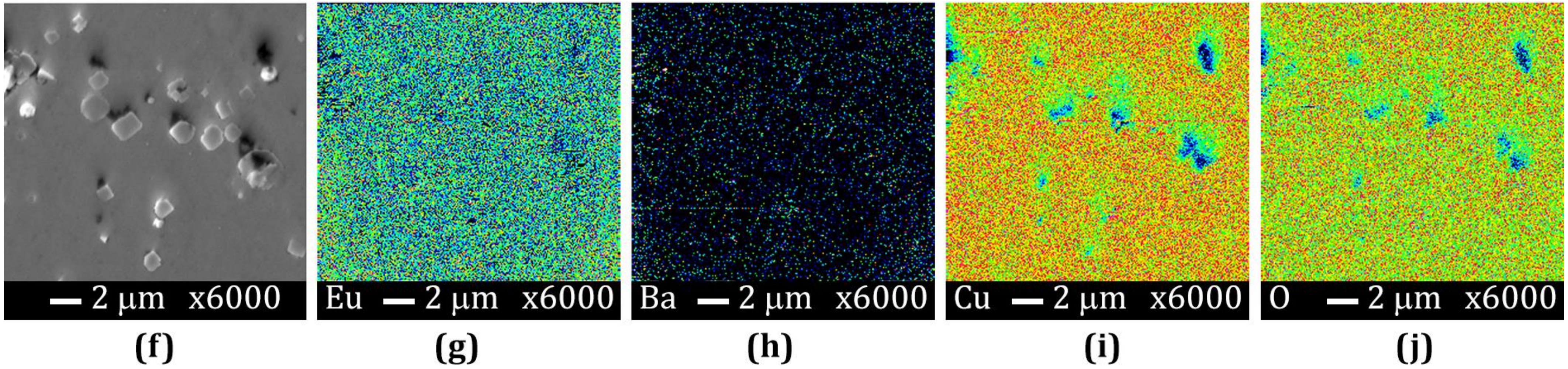
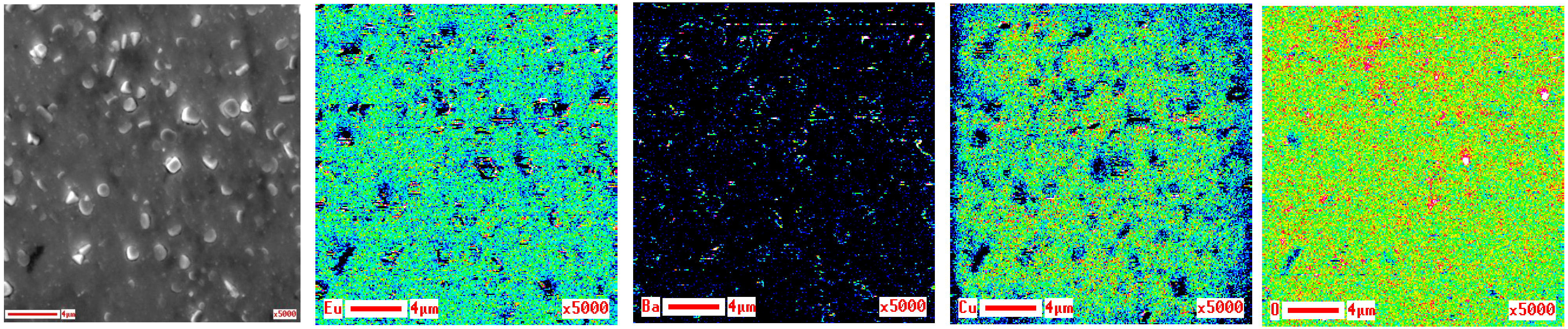
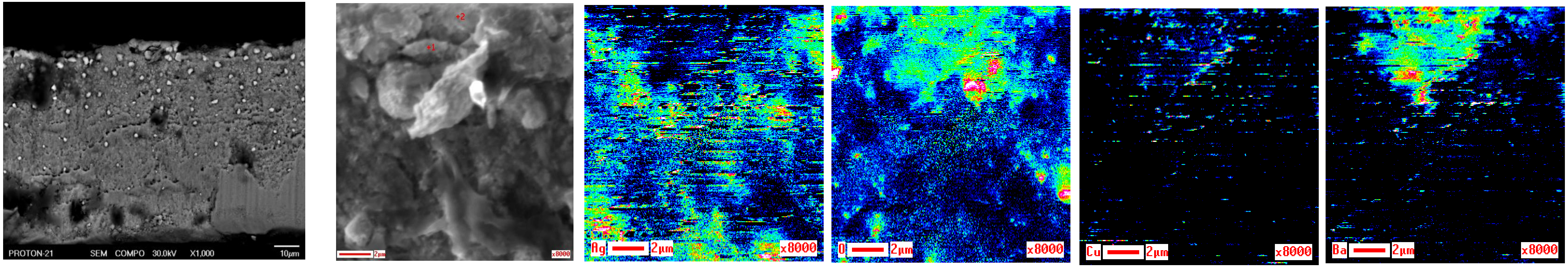



**Figure (a, f) – Images in SEI (secondary electrons) mode of GdBCO_CC and EuBCO+BHO_CC (materials without Cu and Ag layers, which were removed by etching in acids), respectively, and Auger maps of elements distributionover these images: (b-e) – Eu, Ba, Cu, O; (g-j) – Gd, Ba, Cu, O.**

## No 14 EuBCO+BHO_CC (160 bar $O_2$, 800 °C, 3 h)




## No 4 GdBCO_CC (100 bar $O_2$, 600 °C, 3 h)




**S1**

| Elements | Back | Peak | RSF | Intensity | at.% |
|---|---|---|---|---|---|
| C | -77 | 45 | 0,08 | 122 | 12,3 |
| O | -753 | 626 | 0,365 | 1379 | 30,4 |
| Cl | -294 | 257 | 0,7 | 551 | 6,3 |
| Cu | -63 | 41 | 0,21 | 104 | 4,0 |
| Rh | -32 | 39 | 0,5 | 71 | 1,1 |
| Ag | -131 | 164 | 0,78 | 295 | 3,0 |
| Ba | -115 | 116 | 0,085 | 231 | 21,9 |
| Gd | -21 | 44 | 0,025 | 65 | 20,9 |

**S2**

| Elements | Back | Peak | RSF | Intensity | at.% |
|---|---|---|---|---|---|
| C | -136 | 66 | 0,08 | 202 | 14,2 |
| O | -1255 | 1063 | 0,365 | 2318 | 35,7 |
| Cl | -103 | 104 | 0,7 | 207 | 1,7 |
| Cu | -87 | 63 | 0,21 | 150 | 4,0 |
| Rh | -22 | 51 | 0,5 | 73 | 0,8 |
| Ag | -211 | 279 | 0,78 | 490 | 3,5 |
| Ba | -201 | 166 | 0,085 | 367 | 24,3 |
| Gd | -15 | 55 | 0,025 | 70 | 15,8 |

**Table 2. Approximate stoichiometry (according EDS study) and amount of oxygen, at. % ( on 1 atom of Cu estimated by Auger quantitative analysis) in GdBCO and EuDCO+ BHO. The numbering is the same as in Table 1.**

| No | Material | Approximate stoichiometry according SEM EDS | | Amount of oxygen according to Auger analyses, at.% | |
|---|---|---|---|---|---|
| | | Matrix | Inclusion | Matrix | Inclusion |
| 1 | GdBCO_00_CC | $GdBa_{1.8}Cu_{2.6}O_7$ | $GdBa_{1.8}Cu_{2.6}O_{7.8}$ | 60.8 | 62.2 |
| 2 | GdBCO_01_CC | $GdBa_2Cu_{2.8}O_xAg_y$* | Did not estimated | 30.4 | Did not estimated |
| 4 | GdBCO_03_CC | $GdBa_{1.4}Cu_{2.7}O_{8.9}Ag_y$* | Did not estimated | 52.2-58.2 | Did not estimated |
| 10 | EuBCO_00_CC | $EuBa_2Cu_3O_8$ | $EuBa_{2.1}Cu_{2.9}O_{9.6}$ | 59.8 | 60.6 |
| 13 | EuBCO_03_CC | $EuBa_{2.1}Cu_{2.9}O_{7.8}$ | $EuBa_{2.1}Cu_{2.9}O_{8.5}$ | 55.2 | 56.2 |
| 14 | EuBCO_04_CC | $EuBa_2Cu_{2.9}O_{7.1}$ | $EuBa_2Cu_{2.9}O_{8.3}$ | 54.0 | 52.2 |

* The compositions of GdBCO_01_CC and GdBCO_03_CC could not be analyzed properly due to the presence of an Ag top layer; The analysis of the layered structure was carried out from the cut side partially crushed as a result of the cut. When studying GdBCO_00_CC, the Ag layer was removed and the top side was analyzed.

# Conclusions

- Treatment of GdBCO_CC under 100 bar of $O_2$ at 600 °C for 3 h (cooling Mode 1) led to an increase in $J_c$ (77K, 0 T) by 6% and a decrease in the c-parameter of Gd123 to 1.17310 nm, which may be associated not only with overdoping with oxygen, but also with silver diffusion into Gd123.
- No correlations was observed between $J_c$, $T_c$, c-parameter of RE123 (RE=Eu, Gd) and carrier density $n_H$ of EuBCO_CC and GdBCO_CC treated at 300-800 °C, 1-160 bar $O_2$ for 3-12 h.