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Abstract. $REBa_2Cu_3O_{7-x}$ (REBCO; RE = Y, Gd) coated conductor wires are commercially available up to a length of about 1 km. However, for large-scale devices like superconducting coils for high-field magnets several kilometres of a coated conductors are required. Therefore, it is desirable to use joints, which exhibit similar superconducting properties as the coated conductor itself. In this study, we jointed commercial REBCO coated conductors by a two-step procedure. At first, a superconducting soldering solution was developed and deposited on un-stabilized coated conductors via chemical solution deposition. The soldering precursor is based on a Cu-rich solution with a metal cation ratio Y:Ba:Cu of 1:2:4. Secondly, a piece of the coated conductor was delaminated between the superconducting and the buffer layer and used as bridge between two other conductors covered with the soldering layer. Annealing the resulting bridge joint under pressure results in a mechanical stable configuration.

1. Introduction

The $REBa_2Cu_3O_{7-x}$ (REBCO; RE = Y, Gd) coated conductor technology is one of the major routes for the preparation of superconducting wires from high temperature superconductors. There is an increasing interest during the last years to bring such REBCO coated conductors into application, for instance as resistance-free cables for energy transmission, motors, or superconducting coils for high-field magnets. For the application in large-scale devices several kilometers of coated conductors are needed. However, coated conductors are commercially available up to a maximum length of about one kilometer. This length is mainly limited by the production technology. Therefore, jointing techniques for coated conductors are developed to realize even longer wires. State-of-the-art are low resistance Ohmic soldering joints based on lead-tin or silver soldering material [1, 2]. Since the quality of the complete wire depends on the weakest point in the architecture, a fully superconducting joint is desirable for some applications, where any losses need to be avoided in current transport.

Recently, first approaches to realize such superconducting joints have been reported in literature [3-5]. Park et al. [6] presented a zero-resistance lap joint by partial melting of REBCO films in coated conductors. An oxygen annealing process is required afterwards to restore the oxygen stoichiometry in the joint [7]. Therefore, micro holes were drilled with a laser through the complete coated conductor architecture to create artificial diffusion paths for oxygen during annealing, which was performed afterwards for 350 hours in pure oxygen.

Alternatively, Teranishi et al. [8] formed a joint using an additional precursor layer on a REBCO coated conductor prepared by a metal organic deposition (MOD) process. In dependence of the applied jointing

pressure a connected area of up to 35% was reported. The oxygen annealing took 200 hours in pure oxygen flow to restore the correct stoichiometry and resulted in a joint with a superconducting transition temperature $T_{c,0}$ of 82 K and negligible joint resistance, respectively.
Our study used a similar approach and aims to improve the connectivity for the realization of a mechanical stable joint. Therefore, we have developed a Cu-rich $YBa_2Cu_3O_{7-x}$ (YBCO, Y123) precursor solution for a chemical solution deposition (CSD) process. The films prepared by such a CSD solution consist of a superconducting Y 123 phase and an additional Ba-Cu-O phase. The Ba-Cu-O material forms a liquid phase during the jointing process. The second aim of our study is to study approaches to minimize the time for oxygen annealing after the jointing process significantly. Therefore, a piece of a coated conductor was delaminated between the superconducting and the buffer layer in order to test it as a bridge between two other conductors covered with the Cu-rich soldering layer.

2. Experimental procedure

For preparation of the Cu-rich precursor solution yttrium, barium and copper acetate (purity ≥ 99.99%; Alfa Aesar) having a stoichiometric ratio of Y:Ba:Cu = 1:2:4 with respect to the metal cations were dissolved in deionized water and trifluoroacetic acid (99.5+%; Alfa Aesar). The solution was dried and the resulting gel was dissolved in dried methanol (max. 0.003% H_2O; Merck). In order to further reduce the remaining water in the gel, methanol was evaporated and re-filled two more times. The final concentration of 0.25 mol/l of yttrium was adjusted by pure methanol [9, 10, 11]. A standard YBCO precursor solution with Y:Ba:Cu = 1:2:3 was used for comparison. The precursor solution was deposited on (100) oriented lanthanum aluminate ($LaAlO_3$; LAO) single crystal substrates with a size of (10x10x1) mm^3 by spin-coating to study the phase formation. Afterwards, the precursor solution was deposited by dip-coating on unstabilized coated conductor pieces of (4x40) mm^2. After deposition, the precursor films were heat-treated in a quartz tube furnace. This annealing procedure consists of four different steps: (1) the evaporation of residual solvents up to 60°C in dry oxygen atmosphere; (2) a pyrolysis step at up to 400°C in a wet oxygen gas flow to decompose the organic content; (3) the crystallization at T_{max} = 780°C for 60 minutes in humidified nitrogen atmosphere containing 100 ppm O_2 and (4) an oxygenation at 450°C for 2.5 hours in pure oxygen atmosphere. Subsequently, the furnace cooled down naturally. A film thickness of about 600 nm was measured for the crystallized Cu-rich YBCO layers on the coated conductors after two CSD cycles.
A commercially available REBCO-based coated conductor by SuperPower Inc. was used in our study. The coated conductor is composed of a Hastelloy substrate (50 µm), a buffer layer stack (≈ 0.2 µm), the REBCO superconducting film (2 µm). Everything is surrounded by an Ag (≈ 2 µm) and a Cu (20 µm) stabilizing layer. More details regarding the coated conductor fabrication and architecture can be found elsewhere [12]. For the jointing process, the coated conductors was unstabilized by etching the Cu-layer in Ammonium Persulfate (20%; VWR) and the Ag-layer in a mixture of NH_3 (28-30%, Merck), H_2O_2 (35%, Alfa Aesar) and deionized water in the ratio 1:1:1.
The jointing of the coated conductors was realized in a bridge type geometry, with three about one cm long pieces of coated conductors each covered by the Cu-rich YBCO soldering film. The bridge joint was annealed in a tube furnace up to 825°C for 30 minutes under pressure. Therefore, the bridge joint was placed between two pressure plates made from porous silicon carbide. The pressure to the joint was generated by screwing the plates together. In some experiments delaminated coated conductor pieces were taken as bridge in the joint [4, 13]. For delamination, a thermic tension was applied on the coated conductor by using fast temperature changes between liquid nitrogen at 77 K and room temperature several times. The delamination occurred between the REBCO and the buffer layer, resulting in delaminated coated conductor architecture with Cu, Ag and superconducting REBCO layers only.
The microstructure of the different samples covered with a Cu-rich soldering layer as well as of the joints was determined by X-ray diffraction using $\theta/2\theta$-scans (XRD, BRUKER D8 Advance, Co K_α radiation) and by scanning electron microscopy on cross sections prepared with the focused ion beam (FIB) technique (Gemini 1540 XB, Zeiss). A thin platinum film was deposited on top of the REBCO

for the preparation of FIB cuts in order to avoid the deterioration of the REBCO surface by the Ga^{3+} ions.

The critical temperature T_c was measured in a commercial physical property measurement system (PPMS, Quantum Design) by a standard four-probe method. The critical current density J_c at 77 K in self field was measured by an inductive technique (Cryoscan, Theva).

3. Results and discussion

Standard YBCO precursor solutions with a metal cation ratio of Y:Ba:Cu = 1:2:3 result in a superconducting $YBa_2Cu_3O_{7-x}$ (Y123) phase with space group (SG) *Pmmm* after heat treatment as shown in the X-ray pattern in Fig. 1a for the sample prepared on a LAO single crystal. The particular precursor solution for the joint was adjusted to a ratio of Y:Ba:Cu = 1:2:4, resulting in a film containing the Y123 superconducting phase and a barium-copper-oxide (Ba-Cu-O) phase as shown in the X-ray diffraction patterns in Fig. 1b. The highest reflections in both scans are related to the LAO substrate planes and to the (00ℓ) planes of the $YBa_2Cu_3O_{7-x}$ crystal structure. Additional minor reflections appear in the Cu-rich sample at 2θ of 33.1° and 63° representing the $BaCuO_2$ phase with SG *Im*-3*m* und at 2θ of about 41.5° and 69° correlated to a CuO phase (SG *C*/2*c*). In reference [14], the pseudo-binary phase diagram of YBCO is shown with an equilibrium of the Y123 phase and the $BaCuO_2$ / CuO phases up to the eutectic temperature. Above the eutectic temperature $BaCuO_2$ and CuO form a liquid phase with a Ba-Cu-O composition. It was reported that this liquid Ba-Cu-O phase has a significant impact on the successful jointing of top seeded melt grown YBCO [14]. Superconducting properties in the Y123 film and the Cu-rich film on LAO are similar with a $T_{c(50)}$ of 91.5 K.

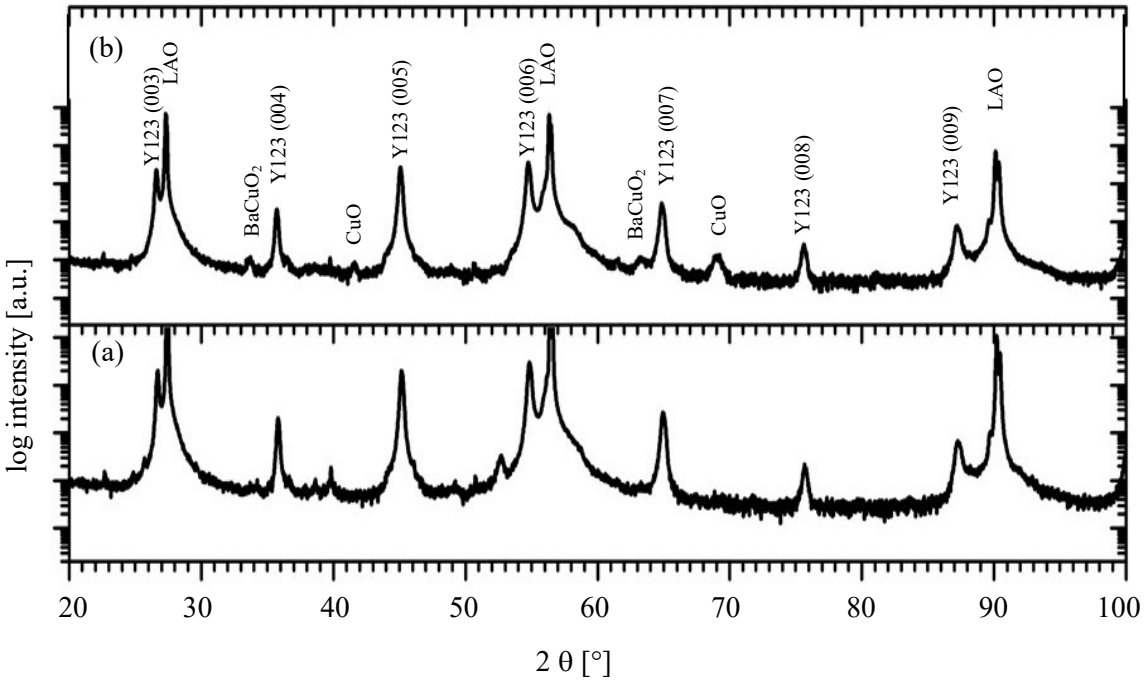

Figure 1. XRD θ - 2θ scans of YBCO films on a LAO single crystal fabricated with a YBCO precursor solution of: (a) a metal cation ratio of Y:Ba:Cu = 1:2:3 and (b) a Cu-rich precursor solution with a metal cation ratio of Y:Ba:Cu = 1:2:4.

The superconducting transition temperature of the unstabilized coated conductor and of the coated conductor covered with the Cu-rich soldering film was measured by a standard four-probe method (Fig 2a). The critical temperature $T_{c(50)}$ of the coated conductor (as received and destabilized by etching) was determined to 92.4 K with a sharp transition into the superconducting phase. An almost unchanged critical temperature $T_{c(50)}$ of 93.2 K was measured after deposition of the Cu-rich soldering film onto the unstabilized coated conductor by dip coating into the corresponding precursor solution and subsequent pyrolysis, crystallization and oxygen annealing.. For simulating the jointing process, the coated conductor with the Cu-rich soldering film was exposed to the jointing temperature of 825 °C for 30 minutes. As expected, the $T_{c(50)}$ is reduced to about 50 K due to the change in the oxygen stoichiometry. After oxygen annealing, the initial T_c value of the coated conductor was recovered to $T_{c(50)}$ = 93.8 K. Similar critical temperatures of the coated conductors before and after the coating and jointing process indicate, that the original REBCO coated conductor properties are not severely influenced by the processes.
Furthermore, the critical current density was inductively measured on Cu-rich YBCO films deposited on single crystal LAO substrates (Fig. 2b). The value of the critical current in the Y123 film is 2.7 MA cm^{-2}. In the Cu-rich YBCO film the critical current density is reduced to 1.1 MA cm^{-2} due to the relatively high content of the Ba-Cu-O phase. However, the reduced J_c value in the Cu-rich YBCO film is not necessarily detrimental as it might be compensated in the jointing process by the size of the overlapping area of the bridge joint in order to carry a similar critical current.

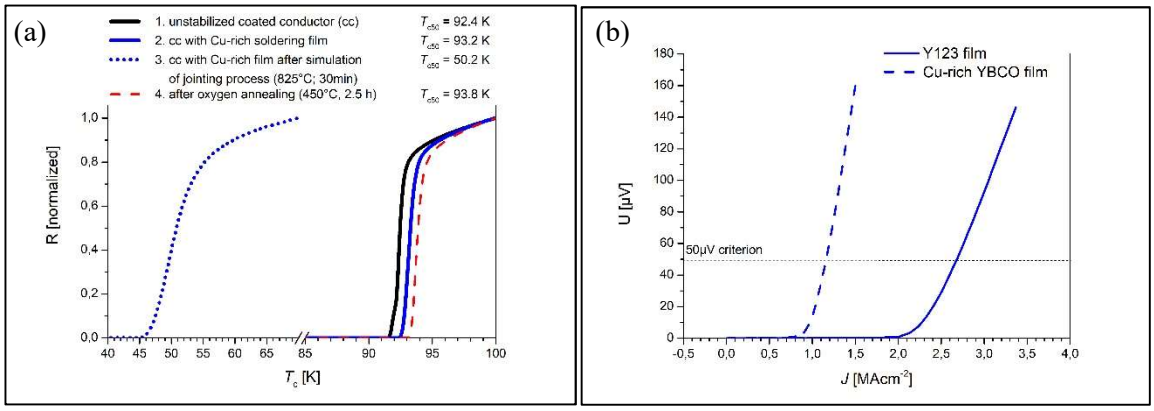

Figure 2. Superconducting measurements of: (a) the critical temperature of 1. unstabilized coated conductor, 2. coated conductor, covered by a Cu-rich YBCO film prepared by CSD, 3. after subsequent exposure to the jointing temperature of 825°C for 30 minutes and 4. after oxygen annealing; (b) critical current density at 77 K (self-field) of a standard Y123 film and a Cu-rich YBCO layer both prepared by CSD on single crystal LAO substrates, respectively.

The jointing of the coated conductors was realized by a bridge joint with two different assemblies: for joint *A* two parallel and one bridge piece of the unstabilized coated conductors, all covered with the Cu-rich soldering film, were assembled. To study the connection in more detail using FIB cuts, the joint was mechanically separated after the jointing process. In this case, the fracture took place at the interface between the buffer layer and the superconductor of the upper conductor and not at the soldered interface indicating a high mechanical stability of the joint itself. For joint *B* a delaminated coated conductor was used as bridge between two parallel coated conductors covered with the Cu-rich soldering film. For the microstructural investigation of the jointed area the copper layer was partially etched. The jointed area is observed in the FIB cross section images (Fig. 3). Figure 3a shows joint *A* with the Hastelloy substrate, buffer layer, the REBCO film and the CSD covered Cu-rich soldering film of the bottom coated conductor followed by the Cu-rich solder and the REBCO film of the upper one. It is visible that the jointing area is inhomogeneous, but it shows still a strong connectivity. In Figure 3b representing joint

B, the jointing area is located between the Cu-rich soldering film of the lower coated conductor and the REBCO film of the delaminated bridge. No gap is visible in the jointing area indicating a significantly improved joint with only a small number of pores.

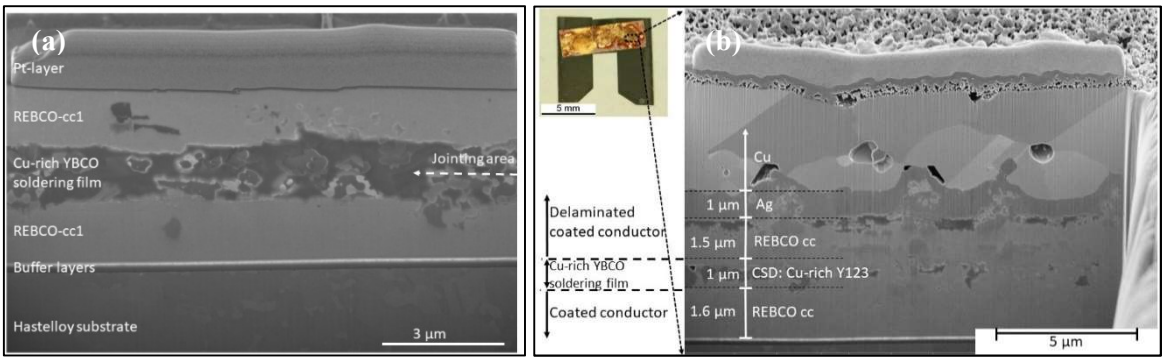

Figure 3. SEM images taken on FIB cross sections: (a) of a joint between two complete coated conductors covered by a Cu-rich soldering film and (b) of a joint between a coated conductor covered by a Cu-rich soldering film and a delaminated coated conductor with remaining copper, silver and REBCO-layers.

During the jointing process at high temperatures of up to 825 °C under reduced oxygen pressure the original oxygen rich superconducting orthorhombic Y123 phase is transformed into a non-superconducting tetragonal Y123 phase due to the loss of oxygen during the annealing procedure. Therefore, an oxygen annealing of the joint is necessary after the forming step. For joint *A* with the coated conductor as bridge, no oxidation was realized during several days of annealing in pure oxygen at 450 °C, i.e. no superconducting behavior was found at 77 K by measuring over the joint. The reason is most probably that the Hastelloy substrate significantly blocks the oxygen diffusion as no diffusion paths are provided. An oxygen annealing of joint *B* with a bridge used from a delaminated coated conductor should theoretically require a much shorter timeframe. However, the annealing of the joint in pure oxygen with an upper copper layer on the bridge results in the formation of thick copper oxide, which is very brittle and mechanically unstable and led to cracks in the complete bridge. This behavior prevented the measurement of the superconducting properties for joint *B*. A solution to this Cu-oxidation problem might be to replace the copper stabilizer by a thicker silver layer. It is known from literature that using silver instead of copper provides permeability for oxygen gas without a chemical reaction at 450 °C [15]. The diffusion rate of oxygen in silver is reported to 1 x 10^{-11} m^2 s^{-1} at 450 °C [16]. Under these conditions, the oxygenation may be completed after about 8 h at 450 °C for a bridge joint with a 20 µm thick silver top layer.

4. Conclusions

A Cu-rich precursor solution has been developed for a chemical solution deposition process to provide a soldering layer for REBCO coated conductors. This layer was deposited on commercial unstabilized coated conductor tapes resulting in a mixture of superconducting Y123 and $BaCuO_2$- and CuO phases. The last two compounds form a liquid phase with a Ba-Cu-O composition during the jointing process at 825 °C. The original superconducting properties of the coated conductor are not influenced by the chemical solution deposition and the jointing process. Two bridge joints were fabricated: *A* – between coated conductors, which were completely covered by the Cu-rich soldering layer and *B* – between a delaminated coated conductor as bridge over unstabilized coated conductors covered with the Cu-rich soldering layer. Due to the presence of the liquid phase in the Cu-rich soldering layer, the microstructure of the joints display a dense jointing area with no gap and only a small number of pores. Nevertheless, the process requires further improvement to achieve fully superconducting joints.

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References
[1] Brittles G D, Mousavi T, Grovenor C R M, Aksoy C and Speller S C 2015 Persistent current joints between technological superconductors *Superconductor Science and Technology 28, 093001.*
[2] Wu X, Wu W, Zhu J, Chang T, ZhaoY, Zhang Z-W, Pan Y, Zuo J, Li Z, Hong Z, and Zhijian J 2017 Electromechanical Performance Study on Silver Diffusion Joints of REBCO Coated Conductors Under Axial Tensile Stress *IEEE Transactions on Applied Superconductivity 27, 6600806.*
[3] Ohki K, Nagaishi T, Kato T, Yokoe D, Hirayama T, Ikuhara Y, Ueno T, Yamagishi K, Takao T, Piao R, Maeda H, Yanagisawa,Y 2017 Fabrication, microstructure and persistent current measurement of an intermediate grown superconducting (iGS) joint between REBCO-coated conductors *Superconductor Science and Technology 30, 115017.*
[4] Jin X, Yanagisawa Y, Maeda H and Takano Y 2015 Development of a superconducting joint between a $GdBa_2Cu_3O_{7-\delta}$-coated conductor and $YBa_2Cu_3O_{7-\delta}$ bulk: towards a superconducting joint between RE (Rare Earth) $Ba_2Cu_3O_{7-\delta}$ coated conductors *Superconductor Science and Technology 28, 075010.*
[5] Hiramatsu K, Teranishi R, Yamada K, Sato Y, Kaneko K 2016 Joint of $REBa_2Cu_3O_{7-\delta}$ Coated Conductors using Metal Organic Deposition *Physics Procedia 81, 109–112.*
[6] Park Y J, Lee M W, Oh Y K and Lee H G 2014 Laser drilling: enhancing superconducting joint of $GdBa_2Cu_3O_{7-\delta}$ coated conductors *Superconductor Science and Technology 27, 085008.*
[7] Eatough M O, Gindley D S, Morosin B and Venturini EL 1987 Orthorhombic-tetragonal phase transition in high-temperature superconductor $YBa_2Cu_3O_7$ *Applied Physics Letters 51, 367–368.*
[8] Teranishi R, Hiramatsu K, Yasuyama S, Miyajima T, Sato Y, Kaneko K, Awaji S, Matsumoto A, Inoue M 2019 Superconducting joint of $GdBa_2Cu_3O_y$ coated conductors by crystallization of additionally deposited precursor layer *IEEE Transactions on Applied Superconductivity, 29, 6602904*
[9] Gupta A, Jagannathan R, Cooper E I, Giess E A, Landman J I and Hussey B W 1988 Superconducting oxide films with high transition temperature prepared from metal trifluoroacetate precursors *Applied Physics Letters 52, 2077–2079*.
[10] Mankiewich P M, Scofield J H, Skocpol W J, Howard R E, Dayem A H and Good E 1987 Reproducible technique for fabrication of thin films of high transition temperature superconductors *Applied Physics Letters 51, 1753–1755.*
[11] Erbe M, Hänisch J, Freudenberg T, Kirchner A, Mönch I, Kaskel S, Schultz L and Holzapfel B 2014 Improved $REBa_2Cu_3O_{7-x}$ (RE = Y, Gd) structure and superconducting properties by addition of acetylacetone in TFA-MOD precursor solutions *Journal of Materials Chemistry A 14, 4932-4944.*
[12] http://www.superpower-inc.com/content/2g-hts-wire
[13] Feng F, Fu Q, Qu T, Mu H, Gu C, Yue Y, Wang L, Yang Z, Han Z and Feng P 2018 Critical current survival in the YBCO superconducting layer of a delaminated coated conductor *Superconductor Science and Technology 31, 045005.*
[14] Cardwell D A, Bradley A D, Babu N H, Kambara M and and Lo W 2002 Processing, microstructure and characterization of artificial joins in top seeded melt grown Y–Ba–Cu–O *Superconductor Science and Technology 15, 639–647.*
[15] Celentano G, Augieri A, Mauretti A, Vannozzi A, Angrisani Armenio A, Galluzzi V, Gaudio S, Mancini A, Rufoloni A, Davoli I, Del Gaudio C and Nanni F 2010 Electrical and Mechanical Characterization of Coated Conductors Lap Joints *IEEE Transactions on Applied*

Superconductivity 20, 1549-1552.

[16] Zhou Z, Ma Y, Han Q and Liu Y 2016 Solubility, permeation, and capturing of impurity oxygen in Au/Ag: A comparative investigation from first-principles *Computational Materials Science 114, 79-85.*