Magnesium diboride on inner wall of copper tube: A test case for superconducting radio frequency cavities

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Superconductor magnesium diboride is considered one of the viable materials to substitute bulk niobium for superconducting radio frequency cavities. Utilizing a MgB_2 coating on the inner wall of a copper cavity will allow operation at higher temperatures (20–25 K) than Nb cavities due to the high transition temperature of MgB_2 (39 K) and the high thermal conductivity of Cu. In this paper, we present results of MgB_2 coating on Cu tubes with similar dimensions to a 3 GHz cavity, as the first step towards coating the actual cavity, using the hybrid physical chemical vapor deposition technique. The results show successful coating of a uniform MgB_2 layer on the inner wall of the Cu tubes with T_c as high as 37 K.

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I. INTRODUCTION

Bulk Nb superconducting radio frequency (SRF) cavities are a well-researched technology and have been successfully deployed in superconducting particle accelerators. However, Nb SRF cavities are quickly approaching their theoretical limits in terms of the highest achievable accelerating gradient (E_{acc}), which is ultimately limited by the thermodynamic critical field (H_c) [1]. Nb has a transition temperature, T_c, of 9.25 K, thus Nb-based SRF cavities typically operate at a temperature of 2 K, which requires pumping liquid He and complex refrigeration, thereby adding substantial cost. Due to these drawbacks of bulk Nb SRF cavities, the search for new materials with higher T_c and better rf performance has been a focus of SRF research. MgB_2 is one of the materials of interest for SRF cavities. It possesses appealing superconducting properties for SRF applications, including high T_c of 39 K [2], low residual resistivity [3], high thermodynamic critical field H_c [4], and absence of weak links at grain boundaries [5]. With these properties, theoretical calculations have predicted a high quality factor (Q) and high E_{acc} for MgB_2 coated SRF cavities [4,6]. In addition, the high T_c of MgB_2 will allow MgB_2 coated SRF cavities to work at higher temperatures above 20 K. MgB_2 coated Cu cavities will have an added advantage from the high thermal conductivity of Cu, which will ensure the enhanced heat transfer from the MgB_2 layer, eventually improving the cavity's resistance to "quenching." This has been observed in Nb coated Cu cavities [7]. MgB_2 coated Cu cavities working at 20–25 K will eliminate the need for liquid He refrigeration [8].

We have grown MgB_2 films on two-inch Cu disks previously using hybrid physical chemical vapor deposition (HPCVD) and tested their superconducting properties at dc and rf frequencies. Growth of MgB_2 on Cu is challenging because Mg and Cu form alloys at elevated temperatures. This problem has been solved by lowering the deposition temperature to around 470 °C [8], where the formation of Mg_2Cu promotes the growth of MgB_2 [9,10]. The films showed high T_c, high critical current density (J_c) and high Q when measured in a cryogenic high-Q hemispheric cavity at 11.4 GHz [8]. Coating of stainless steel dummy 6 and 3 GHz SRF cavities with MgB_2 using HPCVD has been previously demonstrated [11,12]. In this paper, we discuss coating of the inner wall of Cu tubes, which have similar dimensions to a 3 GHz cavity beam tube using a modified HPCVD setup. Deposition conditions for uniform coating of a MgB_2 layer with good superconducting properties have been investigated. The results are a successful first step towards the coating of the actual Cu cavity for testing at 3 GHz.

II. EXPERIMENTAL DETAILS

To test the deposition conditions for coating the inner wall of 3 GHz Cu cavities, Cu tubes with dimensions similar to the 3 GHz rf cavity beam tubes were coated. The unpolished commercial Cu tubes (not oxygen free) have an outer diameter of 1.675 inches, an inner diameter of 1.495 inches, and a length of ~8.5 inches. The Cu tubes

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were split in two for the ease of cleaning before the deposition and characterization after the deposition. They were cleaned in an ultrasonic bath in soap (micro90), the Cu cleaning solution Citranox, and de-ionized water in succession before they were dried with N_2 gas. The two halves were assembled to form a tube during the deposition.

A photo of the HPCVD system for cavity coating is shown in Fig. 1(a) and schematics of the system with the Cu tube at the start and end positions are shown in Figs. 1(b) and 1(c), respectively. The HPCVD system consists of a thermally insulated gas tube from the top to supply the 5% B_2H_6 in H_2 gas mixture and the H_2 carrier gas, a tubular Mg oven from the bottom of the vacuum chamber as the Mg source, and a ~5-inch diameter tubular heater that heats the entire assembly. The tubular heater has two separate parts, with a removable front half, which allows for loading/unloading of the Cu tube or cavity into the system. There is a ~3.5-inch gap between the top of the Mg oven and the opening of the B_2H_6 gas line where B and Mg react to form MgB_2. The B_2H_6 gas line is enclosed in a stainless-steel tube, which can rotate and move vertically (Z direction), and serve as the mounting mechanism for the Cu tube or cavity to be coated. The photo in Fig. 1(a) shows the HPCVD setup with a Cu tube loaded while the front circular heater is removed.

At the start of the deposition, the Cu tube was at the lowest position such that the reaction zone was at the top of the tube [see Fig. 1(b)]. A carrier gas of 5 Torr H_2 was used during the deposition. The temperature of the tubular heater was maintained at around 660 °C and the temperature of the Mg oven was around 600 °C. As discussed in Ref. [8], a Mg-Cu alloy formed on the Cu surface at this stage before the B_2H_6 gas mixture was introduced, which initiated the MgB_2 deposition. The flow rate of the 5% B_2H_6 in H_2 gas mixture was 20 standard cubic centimeters per minute (sccm). The Cu tube was then moved upward with a speed suited for the intended thickness of the MgB_2 coating. In this configuration, the MgB_2 deposition starts from the top and progresses to the bottom. The end position of the system is shown in Fig. 1(c). Throughout the deposition process, the Cu tube was rotated for uniform coating. For ~850 nm thick MgB_2, the Z motion speed was ~0.33 inch per minute and the deposition time was around 30 minutes.

After the deposition, small swatches of the MgB_2 coated tube from different locations on the Cu tube were cut out for characterization. Surface morphology of the swatches was studied with scanning electron microscope (SEM) using a 470 FEI Quanta FEG system. A quantum design physical properties measurement system was used to determine T_c of the samples by magnetic moment (m) versus temperature (T) measurements in a 50 Oe magnetic field. For cross-sectional studies of the samples, focused ion beam (FIB) was used to mill holes in the swatches using an FEI Strata DB235 dual beam system. The thickness of the MgB_2 layer on the cross-sectional sample was measured by SEM, and the deposition rate thus obtained in terms of the Z motion speed was used to determine the thickness of other samples. Two coated tube samples with different MgB_2

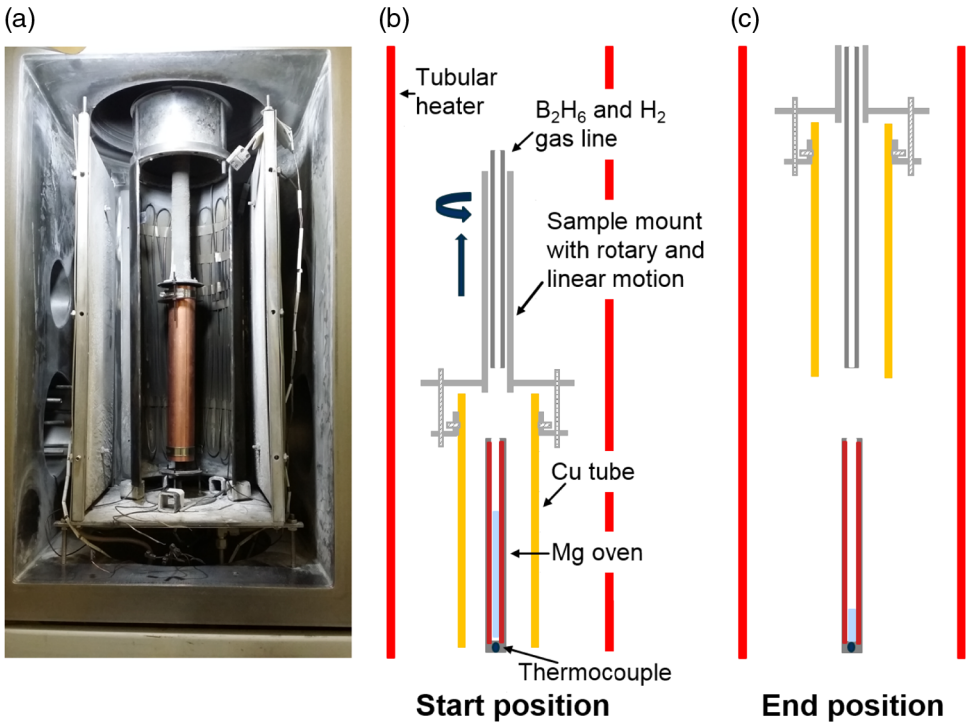

FIG. 1. (a) A photo of the HPCVD system for cavity coating is shown here. (b) The schematic of the system with the Cu tube at the starting position. (c) Schematic of the system with the Cu tube at the end position.

layer thicknesses were characterized by surface morphology and T_c measurements.

III. RESULTS AND DISCUSSIONS

Figure 2(a) shows a photo of the inside wall of a split-half Cu tube after the coating of a ~550 nm MgB_2 film. A range of colors are seen along the length of the tube, which is divided into six regions. Region 1 appears metallic—there is either no coating at all or no MgB_2 coating in this region. From region 2 to region 5, the color gradually changes from dark greenish to purplish. Based on our prior experience, the former is indicative of Mg deficiency and the latter is characteristic of pure MgB_2 films. Irregular light stripes are seen in region 4.

In Figs. 2(b)–2(e), SEM images of the samples from regions 1, 3, 4, and 5 are shown. No MgB_2 was grown in region 1 and the morphology reflects properties of a bare Cu surface with marks of machining. With the gas flow from top to bottom, the top region of the tube experiences gas dynamics different from deep inside the tube, which could result in no coating being present in that region. From region 3 to region 5, well-connected MgB_2 grains can be seen with increasing grain sizes. The variation in the grain size may be the result of changing deposition temperature (cooler at the top than at the bottom of the tube). Cracks in the coating are also observed. With a large mismatch of thermal expansion coefficients (16.5×10^{-6}/K for Cu and 5.4×10^{-6}/K for MgB_2 near room temperature [13]), cracks in the MgB_2 coating on Cu are expected, as the sample experiences contraction when it is cooled from the deposition temperature to room temperature.

Figure 3 shows the zero-field cooled m vs T curves for the samples from region 2 to region 5. All samples showed superconducting transition with T_c ranging from 35 K for region 2 to 37.5 K for region 5.

In Fig. 4(a), a photo of a sample coated with a ~850 nm MgB_2 film is shown. Again, the top region of the tube is not coated. The coating from region 2 to region 6 appears much more uniform than in the thinner ~550 nm sample, particularly in regions 3 and 4. Also absent are the irregular stripes in the thinner sample, although regular stripes perpendicular to the tube axis can be seen, possibly due to the variation of the deposition rate with Z motion. The SEM images of the surface morphology for regions 2, 3, 4 and 5 are shown in Figs. 4(b)–4(e), respectively. Again, well-connected MgB_2 grains are seen with increasing grain sizes from the top to the bottom. It should be noted that no cracks are observed in the ~850 nm sample, although region 5 shows pinholes between the large grains. It appears that the cracks seen in the thinner sample no longer exist in the thicker sample. Regular periodic darker lines can be seen in regions 4 and 5 in Fig. 4(a). We believe that these regular periodic lines are a result of uneven coating area overlap during rotation and Z motion of the Cu tube. Misalignment between the Cu tube, Mg oven and gas line can cause uneven coating.

Cross-sectional samples of the ~850 nm coating on Cu were measured by SEM and the images are shown in Fig. 5. Figures 5(a), 5(b) and 5(c) are for regions 2, 3 and 6, respectively. A thick platinum layer was deposited on top of the sample prior to the FIB milling to protect the MgB_2 layer. The images were taken at a 53.5° angle, which was

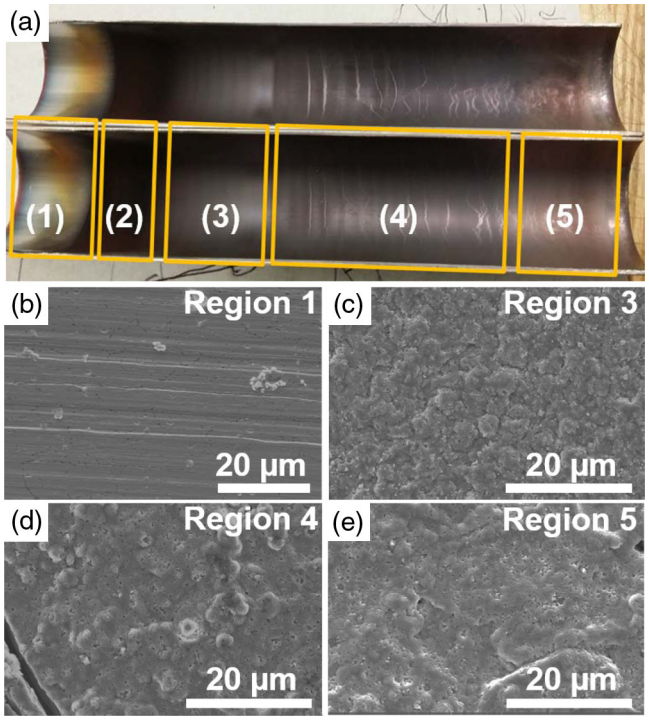

FIG. 2. (a) A photo of ~550 nm thick MgB_2 coated Cu tube halves. (b)–(e) SEM images of MgB_2 film surface on Cu samples cut out from regions 1, 3, 4 and 5, respectively.

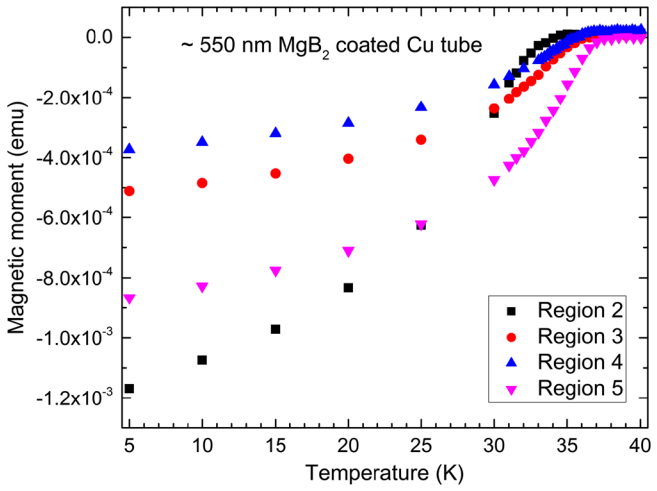

FIG. 3. Magnetic moment vs temperature curves of samples from regions 2, 3, 4 and 5 of a ~550 nm thick MgB_2 coated Cu tube.

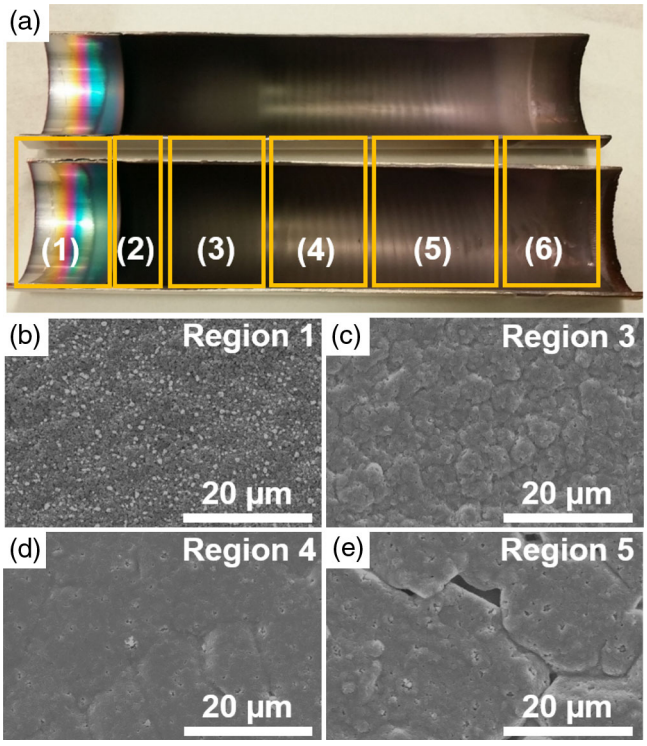

FIG. 4. (a) Photo of ~850 nm thick MgB_2 coated Cu tube halves. (b)–(e) SEM images of MgB_2 film surface on Cu samples cut out from regions 2, 3, 4 and 5, respectively.

accounted for in the thickness calculation. The cross-sectional image from region 2 [Fig. 5(a)] shows an MgB_2 layer intruded by a large number of bright flecks. According to our previous work of MgB_2 coating on Cu disks [8], these flecks are a Mg-Cu alloy appearing in areas where Mg vapor is insufficient during the deposition. It confirms that the top of the tube suffers from Mg deficiency due to the specific gas dynamics in the area. Also visible in Fig. 5(a) is the Mg-Cu alloy layer under the MgB_2 film, similar to that observed in MgB_2 coating on the Cu disk. This layer is not distinguishable in the images for region 3 [Fig. 5(b)] and region 6 [Fig. 5(c)], most likely because the boundary of the alloy layer and the Cu substrate is out of view. Figures 5(b) and 5(c) show dense and conformal growth of the MgB_2 layer with occasional voids near the MgB_2/Cu interface. The blowup of a void in Fig. 5(b) is shown in Fig. 5(d). It is clearly seen that it occurs at a sharp edge on the unpolished Cu surface. It is most likely associated with cracks on thinner MgB_2 coating as seen in Fig. 2, which is then sealed when the film thickness increases. Using these cross-sectional images, the film thickness was determined to be ~850 nm for this sample.

Figure 6 shows the zero-field cooled m vs T curves for the ~850 nm samples from various regions. All the samples showed superconducting transition ~37 K. It should be noted that the samples used in the measurement

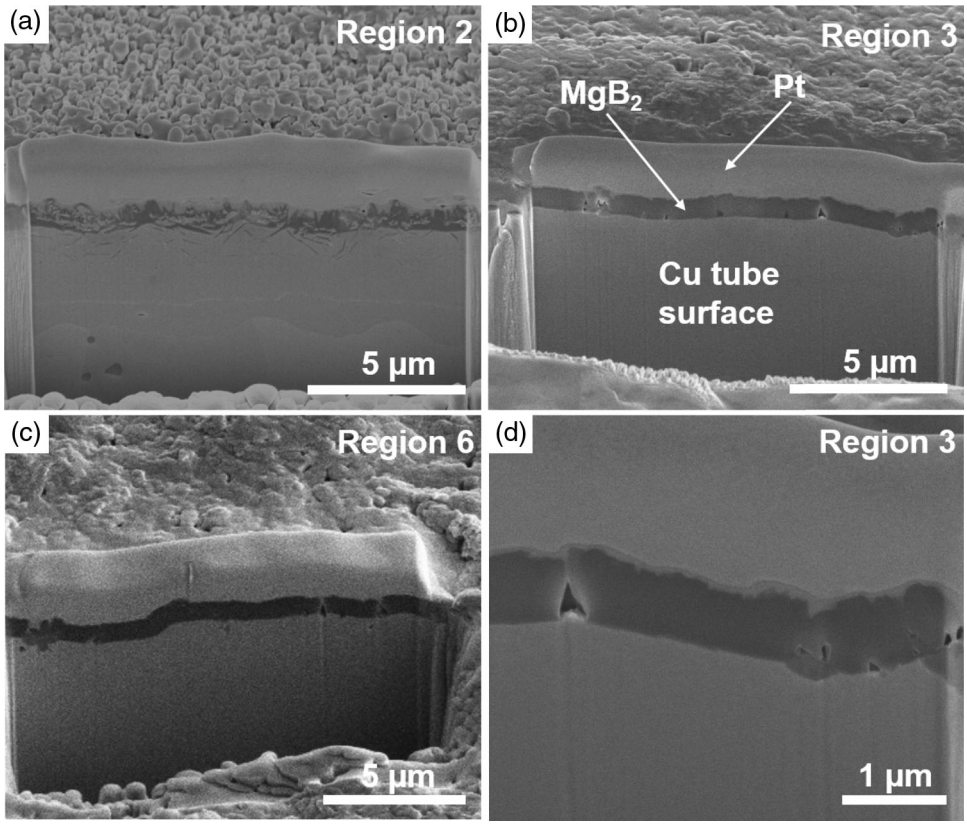

FIG. 5. (a)–(c) Cross section images of samples cut out from regions 2, 3 and 6, respectively, of a ~850 nm thick MgB_2 coated tube. (d) Zoomed in cross section image from region 3.

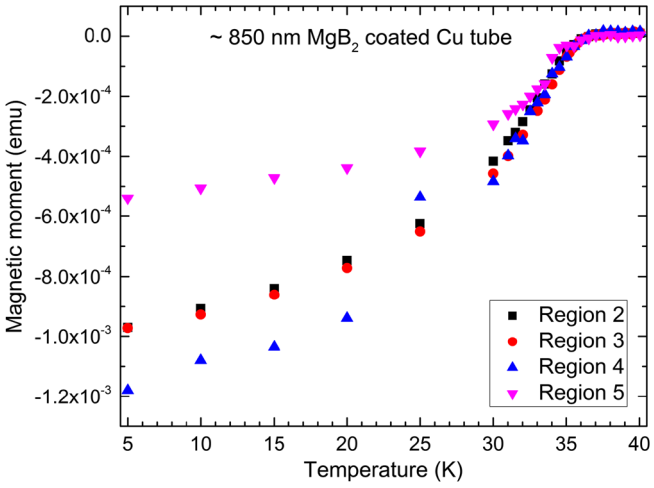

FIG. 6. Magnetic moment vs temperature curves of samples from regions 2, 3, 4 and 5 of a ~850 nm thick MgB_2 coated Cu tube.

were irregularly shaped, therefore the magnetic moment values cannot be directly compared, including those in Fig. 3.

IV. CONCLUSIONS

The results presented here demonstrate that uniform MgB_2 coating with good superconducting properties can be achieved on the inner wall of commercially available copper tubes. The Cu tubes were not oxygen free, thus the outgassing of oxygen may have had detrimental effects on the film properties shown here. This can be alleviated by using oxygen-free copper in future studies. The Cu tubes were not polished, and we show that this may be linked to cracks in thinner films. Preliminary results with polished oxygen-free small Cu disks attached to a Cu tube of the same dimensions as in this work show that the MgB_2 coatings have much smoother surfaces and more complete superconducting transitions in the m vs T curves than the results presented here. While the cracking problem can be solved by using polished Cu tubes in the future, we note that cracks can be sealed in thicker films. Thus, a sufficiently thick MgB_2 coating may be a solution to the possible cracking due to the mismatch in thermal expansion coefficients between MgB_2 and Cu. A remaining issue is the variation of the structure and superconducting properties along the tube axis from one end to the other. The poor coating at either end can be avoided by adding extension tubes at both ends so that only the region with good properties covers the tube or cavity to be coated. The possible variation of deposition temperature with the Z position can be compensated by adjusting the tubular heater temperature while the tube is moving up.

We have successfully coated MgB_2 films on the inner wall of unpolished copper tubes using the HPCVD method. The MgB_2 coatings showed T_c around 37 K over a seven-inch length. A large portion of the coated tube showed dense and uniform coating free from cracks when the thickness of the film was around ~850 nm. The cracks, which were observed in the thinner ~550 nm film, seem to be sealed by the top MgB_2 layer. The results show the feasibility of using the HPCVD method to coat MgB_2 on the inner wall of Cu cavities.

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