In-depth resistance analysis of REBCO tape joints with indium insert and solders

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Abstract. Joints between REBCO (rare-earth barium copper oxide) tapes with low joint resistance are crucial for many superconducting applications. Joining REBCO tapes with indium insert (In-joint) is a promising joining method to fabricate low resistive joints at low temperatures (20–120°C). This study investigated the joining conditions of In-joints such as pickling, surface roughness, joining time, and temperature. The joint resistivity (product of joint resistance and joint area) was successfully reduced to 22–30 nΩ cm² at 77 K in self-field. The constitutive factors of the joint resistivity were analysed separately along with the cross-sectional observations. In this study, the interface resistivity of the REBCO tape was measured as 8.5 nΩ cm² for one REBCO tape by the previously proposed method. The resistivity of the joining interface Cu/In was calculated as <3 nΩ cm² by subtracting the other resistivities from the entire joint resistivity. This result reveals the lower limit of the joint resistivity: the sum of the resistivity (nΩ cm²) of indium (measurable by thickness), the resistivity of Cu/In (<3nΩ cm²), and the interface resistivity of the REBCO tape (measurable beforehand). Furthermore, we demonstrated a lower and less varied joint resistivity of In-joints than those of the soldered joints.

1. Introduction
Joints between REBCO tapes with low joint resistance are crucial for various superconducting applications such as power cables for railway systems [1] and fusion magnets [2]. For years, the authors of Tohoku University have been developing the joining method with indium insert (In-joint) [2-4]. The joint resistivity (product of joint resistance and joint area) of In-joints ranges 25–60 nΩ cm² [4] at 77 K in self-field, which is smaller than those of the soldered joints (30–1000 nΩ cm² [5, 6]). In order to know how the joint resistivity can be reduced, the resistance purely due to joining has to be evaluated because the joint resistivity itself depends on the interface resistivity inherent to REBCO tapes [5]. Therefore, in this study, we separately evaluate the resistance factors of the joint resistivity, including the resistivity at the joining interface and the interface resistivity of the REBCO tape.

The objective of this study is to further reduce the joint resistivity of In-joints by investigating the joining conditions such as pickling, surface roughness, joining time, and temperature. The resistance factors of the joint resistivity were separately evaluated for a reason mentioned above. The cross-sections of the In-joints were also observed to discuss what reduced the joint resistivity. Furthermore, the joint resistivity of In-joints was compared with those of soldered (In52Sn48 and Pb37Sn63) joints to demonstrate the excellent performance of In-joints.
2. Method

2.1. Investigation of joining conditions

In this study, copper-stabilized 4-mm-wide YBCO tapes (SCS4050-AP, $I_c > 91$ A at 77 K in self-field, SuperPower Inc., Schenectady, NY, USA) and 100-µm-thick indium foil were used for the In-joints. The tape has a layer structure of Cu (20 µm) / Ag (2 µm) / buffer layer (~1 µm) / YBCO (1 µm) / Hastelloy-C276 (50 µm) / Ag (2 µm) / Cu (20 µm). In-joint were fabricated in the procedure shown in Figure 1(a). The tapes were set in a face to face manner with indium insert in a jig and joined by using a pressing machine shown in Figure 1(b). The joining pressure was fixed at ~90 MPa because the joint resistivity does not change with the applied pressure over 50 MPa [4]. All the samples were joined with overlapped lengths around 7 mm.

We conducted three experiments (Ex. 1–3) to find useful parameters for resistance reduction. In-joints were fabricated with different joining conditions in the experiments. The conditions of pickling, surface roughness/joining time, and joining temperature were changed in Ex. 1, 2, and 3, respectively. Table 1 describes the details of the joining conditions in Ex. 1–3. For pickling, a commercial flux (SUSSOL-F, Hakko corp., Osaka, Japan) containing ZnCl (35–45%) and NH$_3$Cl (<10%) was used for the Cu surface of the REBCO tapes, and 10% HCl was used for indium foil.

After joining the tapes, the joint resistance was measured at 77 K in self-field by the four-probe method with a current up to 100 A, and the critical current was confirmed not degraded. Additionally, three In-joint samples fabricated in Ex. 2 were heated at 95°C for three or four days in an electric furnace with an applied pressure of 29 MPa, and the joint resistivity before and after heating was measured.

2.2. Cross-sectional observation and joint resistivity evaluation

The cross-sectional of the In-joint fabricated by the previous joining process [4] (without pickling) and the sample fabricated in Ex. 2 (with pickling) were observed with the scanning electron microscope (SEM) and energy dispersive X-ray spectroscopy (EDS). The samples for observation were prepared by cutting and milling with an Ar ion beam at −90°C or −100°C to avoid the deformation of soft indium.

The joint resistivity $r_{\text{joint}}$ (the product of joint resistance and joining area) was separately evaluated from each resistance factor, as shown in the following equation:

$$r_{\text{joint}} = 2 \left(r_{\text{Cu}} + r_{\text{Ag}} + r_{\text{Cu}/\text{Ag}} + r_{\text{Ag}/\text{REBCO}} \right) + 2r_{\text{Cu}/\text{In}} + r_{\text{In}} \tag{1}$$

where $r_{\text{Cu}}$ and $r_{\text{Ag}}$ are the resistivities ($\Omega \cdot \text{cm}^2$) of Cu and Ag layers of a REBCO tape. $r_{\text{Cu}/\text{Ag}}$ and $r_{\text{Ag}/\text{REBCO}}$ are the interface resistivity, $r_{\text{In}}$ is the resistivity ($\Omega \cdot \text{cm}^2$) of In, and $r_{\text{Cu}/\text{In}}$ is the resistivity at the joining interface Cu/In. We did not consider $r_{\text{Cu}}$ and $r_{\text{Ag}}$ because they are negligibly small (~0.1 $\Omega \cdot \text{cm}^2$). As for $r_{\text{Cu}/\text{Ag}}$ and $r_{\text{Ag}/\text{REBCO}}$, we measured the sum of them as 8.5 $\Omega \cdot \text{cm}^2$ per one REBCO tape by using the contact-probing current transfer length method [7]. $r_{\text{In}}$ is the product of the volume resistivity of indium (18.8 $\Omega \cdot \text{m}$, measured by the four probe method at 77 K) and its thickness measured with a micrometer or by SEM observation. $r_{\text{Cu}/\text{In}}$ is calculated as the difference between the joint resistivity and the other resistance factors.

2.3. Comparison with soldered joints

Three samples each of InSn (In52Sn48) and PbSn (Pb37Sn63) soldered joints were fabricated to compare the joint resistivity with that of In-joints. We pressed the solder into a foil-shape before joining and then inserted it between the connected tapes with flux (SUSSOL-F) applied on tape surfaces. We adopted this process because it is superior to the pre-tinning process for low joint resistance [6]. The joining temperatures of InSn and PbSn joints were 140°C and 190°C [8], respectively. After reaching the joining temperatures, a pressure around 10 MPa was applied and held until the jig cooled down to ~70°C.
3. Results and Discussion

3.1. Investigation of joining conditions

Figure 2 (a)–(c) show the results of Ex. 1–3, respectively. Figure 2 (a) shows the joint resistivity of the samples fabricated with different pickling conditions described in Table 1. Two samples were prepared for each condition. Figure 2 (a) indicates that the joint resistivity decreased by ~10 nΩcm² by pickling REBCO tapes and In, compared to without pickling (previous process). Pickling the tape reduced the joint resistivity because flux containing NH₃Cl and ZnCl can remove copper oxide on the tape surface. However, the effect of pickling In was minor, implying that In oxide can be destroyed mainly by the deformation of In during pressing. Figure 2 (b) shows the joint resistivity of the samples made with different conditions of surface polishing and joining time. The joint resistivities of all the samples were almost the same around 30 nΩcm², meaning that these two parameters do not influence the joint resistivity under the condition of the pickling process.

Figure 2 (c) shows the details of the joint resistivity of the samples joined at different temperatures. In this figure, \( r_{\text{In}} \) and \( r_{\text{Cu/In}} \) have an error of ~1 nΩcm² because the thicknesses of indium were measured with a micrometer that has an error of ~5 µm. Generally, \( r_{\text{In}} \) decreased with the increase of the joining temperature because indium gets softer and thinner (easily pushed outside the joint area) at higher temperatures. Note that minimizing \( r_{\text{In}} \) is not essential at very low temperatures where the volume resistivity of indium is negligibly small. Also, in Figure 2 (c), \( r_{\text{Cu/In}} \) was sufficiently small (<3 nΩcm²) and did not change over the different temperatures in a short joining time of 15 min. Similarly, the joint resistivity after heating at 95°C for three or four days did not drastically change, as shown in Figure 2 (d). In short, pickling and joining temperature are useful, but surface roughness and joining time are not to reduce the joint resistivity.

| Table 1. The details of experiments Ex.1–3 |
|-------------------------------------------|
| Ex. 1 | Ex. 2 | Ex. 3 |
|-------|-------|-------|
| **Surface Polish** | #240 | #240–#1500 | #1500 |
| Pickling | REBCO tape & In, ReBCO tape only, In only, None (previous process [4]) | REBCO tape & In, Alumina polishing agent (#3000) | REBCO tape & In |
| Time | 15 min. | 5-90 min. | 15 min. |
| Temperature | Room temperature (RT) | RT | RT–120°C |
| Result | Figure 2 (a) | Figure 2 (b) | Figure 2 (c) |

*# is the grid sizes of the abrasive papers or polishing agent specified in Japanese Industrial Standard (JIS)
Figure 2. Relationships between the joint resistivity and (a) pickling conditions, (b) joining time and surface roughness, (c) joining temperature, and (d) long heat treatment. The details of the joining conditions are described in Table 1. In (c), $r_{\text{In}}$ and $r_{\text{Cu/In}}$ have an error of ~1 n$\Omega$cm$^2$ because of the measurement error of ~5 µm by a micrometer. In (d), Sample 1 was heated at 95°C for 96 hours. Sample 2 and 3 were heated for 68 hours. These samples are marked in (b).

3.2. Cross-sectional observation and joint resistivity evaluation

To discuss the effect of the pickling process, we observed the cross-sections of the samples with and without pickling. Figure 3 shows the cross-sectional SEM images and EDS elemental maps of the sample without pickling. This sample was fabricated by the previous process [4], in which the REBCO tapes were joined at room temperature after surface polishing by a #240 abrasive paper. At the joining interface, we observed an intermetallic compound CuIn$_2$, which forms below 148°C [9]. Also, numerous voids smaller than 1 µm generated in the In region near the joining interface probably because of uneven CuIn$_2$ formation accompanied by volume shrinkage. This volume shrinkage is calculated as 7.15% by the following equation:

$$\Delta V = \frac{V_{\text{CuIn}_2} - V_{\text{Cu+2In}}}{V_{\text{Cu+2In}}} = \frac{m_{\text{Cu}} + 2m_{\text{In}}}{\rho_{\text{Cu}} + 2\rho_{\text{In}}} - 1 = \frac{63.5 + 2 \times 114.8}{8.2} - 1 = -0.0715$$ (2)
where \( V_{\text{CuIn}_2} \) and \( V_{\text{Cu}^{+2}\text{In}} \) are the molar volumes of CuIn\(_2\) and Cu\(^{+2}\)In, \( m_{\text{Cu}} \) and \( m_{\text{In}} \) are the molar masses of Cu and In. The densities of CuIn\(_2\), Cu, In are \( \rho_{\text{CuIn}_2} = 8.2 \) \( \text{g/cm}^3 \), \( \rho_{\text{Cu}} = 7.31 \), and \( \rho_{\text{In}} = 8.96 \) \( \text{g/cm}^3 \) at room temperature. Figure 3(b) describes the details of the joint resistivity of the observed sample. The joint resistivity was mainly shared by the interface resistivity \( r_{\text{Cu/Ag}} + r_{\text{Ag/REBCO}} \) (17 n\( \Omega \)cm\(^2\)), the In resistivity \( r_{\text{In}} \) (16.9 n\( \Omega \)cm\(^2\)) and the joining interface resistivity \( r_{\text{Cu/In}} \) (6.5 n\( \Omega \)cm\(^2\)) that includes the resistivity of CuIn\(_2\). Although the growth of CuIn\(_2\) can increase \( r_{\text{Cu/In}} \), the joint resistivity after long heating did not drastically change as already shown in Figure 2 (d). The volume resistivity of CuIn\(_2\) will be discussed in future study.

Subsequently, we observed the cross-section of the pickled sample marked in Figure 2 (b). In this sample, the CuIn\(_2\) layer formed thickly and uniformly probably because the pickling process decreased Cu oxides that hindered CuIn\(_2\) formation. Besides, the voids near the joining interface decreased considerably compared to those in the sample without pickling (Figure 3(a)). Figure 4(b) shows the detail of the joint resistivity of the pickled sample. The resistivity at joining interface \( r_{\text{Cu/In}} \) was evaluated as <1 n\( \Omega \)cm\(^2\). This significant reduction of \( r_{\text{Cu/In}} \) can be attributed to the decrease of the voids and the uniform formation of CuIn\(_2\) in this pickled sample. Therefore, we conclude that the pickling process sufficiently reduced the resistivity of In-joints.

![Figure 3](image1.png)  
**Figure 3.** (a) SEM images and EDS elemental maps, (b) The details of the joint resistivity of the sample fabricated by the previous process [4]. In the previous process, the tapes were joined at room temperature after surface polishing by a #240 abrasive paper without pickling.

![Figure 4](image2.png)  
**Figure 4.** (a) SEM images and EDS elemental maps, (b) The details of the joint resistivity of the sample with the pickling process. This sample was fabricated in Ex. 2 with surface polished by #1500 and a joining time of 5 min.
3.3. Comparison with soldered joints

Figure 5 compares the joint resistivities of In-joints, InSn joints, and PbSn joints. The joint resistivity of In-joints showed smaller and less fluctuated values than those of soldered joints. Besides, In-joints can be fabricated at lower temperatures of 20–120°C, which reduces the joining time and the risk of $I_c$ degradation of the REBCO tape compared to the soldered joints.

![Figure 5. Joint resistivity of In-joints and soldered joints. In-joints are the samples in Figure 2 (c).](image)

4. Conclusion

This study investigated the joining conditions of In-joints with the analysis of the constitutive factors of the joint resistivity and the cross-sectional observations. Mainly by pickling the Cu surface of the REBCO tape, we successfully reduced the joint resistivity to 22–30 nΩcm$^2$ at 77 K in self-field with joining temperatures of 20–120°C. The joining temperature contributed to reducing the resistance (thickness) of In. However, joining time and surface roughness did not affect the joint resistivity.

The constitutive factors of the joint resistivity were separately evaluated to find that the joint resistivity has been reduced sufficiently. The interface resistivity was measured as 17 nΩcm$^2$ for two REBCO tapes and the resistivity at the interface Cu/In was calculated as <$3$ nΩcm$^2$ with small fluctuations. This result reveals the lower limit of the joint resistivity: the sum of the resistivity (nΩcm$^2$) of indium (measurable from thickness), the resistivity of Cu/In (<3nΩcm$^2$), and the interface resistivity of REBCO tapes (measurable from [7]). The cross-sectional observations revealed that CuIn$_2$ forms at the joining interface but not increases the joint resistivity severely. The volume resistivity of CuIn$_2$ will be examined in future study.

Finally, we compared the joint resistivity of In-joints with those of InSn and PbSn joints. The comparison indicated that In-joints have a reproducibly low joint resistivity by a simple process and lower joining temperatures than those of the soldered joints.

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