Jointing of coated conductors by using nano-particle metal pastes

Tsuyoshi Nakanishi\textsuperscript{a,b}, Takato Machi\textsuperscript{a,b}\*, Teruo Izumi\textsuperscript{a,b}, Ryo Teranishi\textsuperscript{a,c}, Tomohiro Kato\textsuperscript{d}, Takeharu Kato\textsuperscript{d} and Tsukasa Hiyama\textsuperscript{d}

\textsuperscript{a}iSTERA, KSP R&D A-9F, 3-2-1, Sakado, Takatsu-ku, Kawasaki, Kanagawa, 213-0012, Japan
\textsuperscript{b}ISTEC-SRL, KSP R&D A-9F, 3-2-1, Sakado, Takatsu-ku, Kawasaki, Kanagawa, 213-0012, Japan
\textsuperscript{c} Kyusyu, University, 744 Motooka Nishi-ku Fukuoka 819-0395 Japan
\textsuperscript{d} Japan Fine Ceramics Center, 2-4-1 Mutsuno, Atsuta, Nagoya, 456-8587, Japan

Abstract

Development of a jointing technique of coated conductors is important for all applications, such as superconducting magnets, cables, etc. Low resistance jointing techniques by means of silver diffusion \[1\] and for superconducting joints\[2\] have been reported so far. Since these processes were carried out at higher temperatures than the O\textsubscript{2} annealing temperature for appropriate carrier doping to the REBa\textsubscript{2}Cu\textsubscript{3}O\textsubscript{7-\textgamma} (REBCO) crystals and resulted in oxygen deficiency in the REBCO crystals, long time O\textsubscript{2} annealing was required for compensation of this oxygen deficiency. Because the long time and high temperature post annealing is an inappropriate process as on-site technology, solder jointing technology has been widely accepted, in general, for practical applications. However, the resistance of the solder joint is 50 - 100 n\textohm, and then the Joule heat generation in the joint region is a serious problem and must be solved. Consequently, we have studied a new jointing technique by using the pastes containing of silver or gold nano-particles. Because the \textit{I}\textsubscript{c} value of GdBCO was deteriorated with higher temperature heat treatment, we have tried to develop a jointing technology with the low temperature (below 200\textdegreeC). We used the nano-particle metal pastes (~5 nm) which contained dispersants around the chemically active surface of nano-particles and dissociates at low temperatures and achieved the low resistance joint (~ 3n\textohm, 10 x 160mm\textsuperscript{2}, 77 K) as well as no \textit{I}\textsubscript{c} degradation without O\textsubscript{2} post annealing.

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* Corresponding author. Tel.: +81-44-850-1616; fax: +81-44-850-1613.
E-mail address: machi@istec.or.jp
1. Introduction

In order to promote the applications of REBCO coated conductors (CCs), it is better that the jointing resistance should be as low as possible. At present, the solder jointing is frequently used for connecting CCs. The tensile strength of the solder joint in the case of MOCVD CCs was reported by Sugano[3]. From their results, one can recognized the maximum applied load of from around 700 to 800 N. On the other hand, a diffusion jointing technique was developed in our laboratory as the previous work[1]. In the case of diffusion jointing of silver stabilizing layers of MOCVD CCs in a face to face manner, the tensile strength is slightly higher than the solder joint such as about 1 kN. Therefore, we set the tensile strength to be 1 kN as a goal.

The resistance of diffusion joint is 6 to 10 nΩ. Although this value is smaller than that of the solder joint of 50 - 100nΩ, this process included the long post O₂ annealing of about 12 hours. Therefore, this process was not used for the field of application. The superconducting joint was reported by the Korean group[2]. Although it is a very attractive process, the very long O₂ annealing of 2 weeks is needed. Consequently, it is difficult to be used for the fields of applications.

About the solder joint, the strong point is a handy process, although the resistance of the jointing is higher than other techniques. Therefore we have started to develop a new jointing technique to satisfy the following characteristics. At first, the new technique must be handy and needless of the post-O₂ annealing process. Secondly, the resistance of jointing is less than 5 nΩ which was lower than that of diffusion jointing of silver. Thirdly, the tensile strength is higher than 1kN. In order to achieve these requirements, the usage of the nano-particle metal pastes has been tried and investigated.

The nano-particle metal pastes have been used for the low temperature patterning, for example on the plastic tapes. Therefore, it is thought the heat treatment can be done in the low temperatures much lower than the orthorhombic- tetragonal transition temperature of the REBCO superconductors, that is to say that at the temperature does not occur the oxygen deficiency. Accordingly, the post O₂ annealing is thought to be unnecessary.

Nano-particle has very active surface and then there is a possibility to achieve the jointing at low temperature. The surface of nano-particles is covered up by the dispersing agent in order to prevent self-agglomeration. Therefore, the selection of the kinds of dispersing agent (dispersant) is technology to become the key to this process. Firstly, we tried to joint the CCs by using Au nano-paste. Secondary, we tried to joint by using Ag nano-paste. The surface of silver particle is easily affected by oxygen diffusion at low temperatures, and there is a difference between the two types of the pastes. In addition, the nano-paste jointing technique has the advantage of lead free process taking into environmental consideration. In this paper, we will report the results of both cases.

2. Experimental

We used the Au nano-paste (AU NANOMETAL, ULVAC Inc.) and Ag nano-paste (L-Ag1 TeH, ULVAC Inc.). The particle size of nano-paste was about 5 nm in diameter. We used GdBCO coated conductors for specimens with a stabilizing Ag layer of 5µm in thickness.

Firstly, The nano-paste was painted the Ag surfaces of the two specimens with a brush. Secondary, The two specimens were jointed in a face-to-face manner. Thirdly, a set of the specimens was fixed each other by pressing with metal plates and a screw in every 1cm length, where the jointed area was 1 cm width and 8 cm length (8 cm² area size). The heat-treatment was applied using a constant temperature furnace in the air. The temperature was changed from 100 to 250 °C. The holding time for heat treatment at each temperature is 1 hour. In order to confirm the area size dependence of jointing resistance, it was fabricated the jointed specimens with 16 cm² area size.

The evaluations of the specimens include I-V characteristics measurement in the liquid nitrogen, tensile strength test in the room temperature, and cross-sectional SEM observation.
3. Results and discussions

Figure 1 shows the I-V curves between the two specimens jointed by the Au nano-particle paste. The closed symbols(♦) represent a typical I-V curve measured at 77K in the self-field of coated conductor before jointing. Open triangles are the result of the specimen with the case of 150 °C jointing. From the slopes of the linear portion in the curves, we were able to estimate the resistance of the respective jointing. In the case of 250 °C, the I_c value with a criterion of 1 μV/cm decreased by jointing, which might be explained by oxygen deficiency of the superconducting crystals caused by oxygen diffusion at the higher temperature from the superconducting layer to the Ag stabilizing layer and to the nano-pastes. Consequently, it is thought the oxygen deficiency of the superconducting crystals might occur with the higher temperature jointing than 250 °C.

The tensile strength was measured with definition of the yield point where the measured strain deviated from the linear increase with increasing the load. In the case with the 150 °C jointing, the tensile strength is about 1.2 kN. If the tensile load to the specimen was less than the yield point strength, no change in the I-V curves was observed between the specimens before and after the tensile test. It was found from the surface observation of the destructed specimens that the delamination took place at the interface between the REBCO layer and the silver stabilizing layer, not at the joints of the silver layers by the pastes. Therefore, it could be expected that the delamination strength of the joint by nano-pastes was higher than that of CC itself.

From the results of I-V properties, the jointing temperature dependence on the jointing resistance was obtained. It is clear the resistance decreased with decreasing the temperature of jointing, and the very low resistance of 5nΩ for the goal was almost attained (The measured resistance with the 150 °C jointing was 5.69nΩ). The jointing temperature dependence on the tensile strength of the specimen with a joint was also measured as mentioned above and the results are shown with the broken curve of closed square symbols in Fig. 2. The result indicates that the specimens jointed at the higher temperature than 150 °C have the higher tensile strengths exceeding 1kN. Consequently, it could be concluded that the appropriate jointing temperature was 150 °C in order to satisfy both the low jointing resistance as well as high tensile strength of the joint.
The jointing area dependence on the resistance was investigated. The results exhibited that the resistivity of the jointing was almost the same value even though the areas were changed. Therefore, as the jointing resistance could be decreased with increasing the jointing area, we succeeded in attaining $3n\Omega$ of the jointing resistance by increasing the area to $16\ \text{cm}^2$. This value of the resistance was lower than that of $5n\Omega$ required for applications as mentioned in the section of introduction. The thickness of the jointed region between the Au layers was $1.9\ \mu\text{m}$ measured from the cross-sectional SEM image as shown in Fig. 3.

In the case of Ag nano-paste, because the surface area of nano-particle is large, if a little oxygen exists on the surface of the primary nano-particle of silver, there is a possibility of the obstruction of the electric currents. Therefore, we paid special attention to the dispersant which protects the surfaces of the Ag nano-particles from the oxidation and self-agglomeration. The dispersant with the low dissociation temperature was selected in this study. Fig. 4 shows the result of Ag nano-paste jointing. Although jointing with Ag nano-paste could not be successfully obtained at the low temperature of $100^\circ\text{C}$, the specimen jointed at $150^\circ\text{C}$ exhibited similar values of the jointing resistance and the tensile strength to the values of the specimen jointed with the Au nano-pastes. Therefore, it was confirmed that Ag nano-paste jointing can be also used for connecting CCs.

The thickness of the Ag nano-paste region was about $0.5\ \mu\text{m}$ which was estimated from the cross-sectional SEM images of the jointing region. Although some voids existed in the jointing region, the resistance exhibited almost the same value of the specimen jointed with Au nano-pastes, which suggests, in other words, the possibility of further improvement of the jointing performance.

### 4. Conclusion

The most important requirements for the jointing of CCs are handy and with free of post-$\text{O}_2$ annealing. We succeeded in development of a new method for jointing of coated conductors by using nano-metal pastes. By using Au nano-pastes, we could achieve the low temperature jointing in the air within 1hour. The jointing resistance was down to $3n\Omega$ with the jointing area of $16\ \text{cm}^2$. And we attained more than 1kN of the tensile strength in the specimen jointed at $150^\circ\text{C}$, at the same time. All the requirements, including handy together with low resistance and higher tensile strength of the jointing technology for application, were achieved by using Au as well as Ag nano-particle pastes. This new jointing method can be applied in the air and at $150^\circ\text{C}$ of a reasonably low temperature. The resistance obtained was low, and the tensile strength was over 1 kN. It is highly recommended to use this new jointing method for the fields of applications of REBCO coated conductors.

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