Nb$_3$Sn multifilamentary superconductors fabricated through a diffusion reaction between Nb and Ag-Sn alloys

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Abstract. Ag-Sn alloy is very attractive for fabricating Nb$_3$Sn wires through diffusion reaction, because Ag-based alloy including a large amount of Sn is ductile. Therefore we investigated the Nb$_3$Sn formation through the diffusion reaction between Nb and Ag-Sn alloys. With using Ag-9 at% Sn fcc phase, Ag-12 at% Sn $\zeta$ phase, and Ag-24 at% Sn $\epsilon$ phase alloys, we fabricated single-core, 200-core, and 40000-core composite wires with Nb matrix and Ag-Sn alloy cores. With increase of Sn content in Ag-Sn alloys, the obtained superconducting properties of heat treated composite wires were improved. $T_c$ and $B_{c2}(4.2 \text{ K})$ for the Nb/Ag-Sn are similar to those for the Nb/Cu-Sn. However, the $I_c$ values are relatively small, due to the formed very thin Nb$_3$Sn layers. Ag is apparently not so effective to increase the formation rate of Nb$_3$Sn layer as Cu. We obtained very interesting results by making the 200-core and 40000-core wires, which show the improved $T_c$ and $B_{c2}(4.2 \text{ K})$ by 0.5-1 K and 2-5 T, respectively. From 10 to 500 times larger $I_c(4.2 \text{ K})$ were also obtained for the multifilamentary wires.

1. Introduction

Cu is well known to accelerate the formation rate of Nb$_3$Sn in diffusion reaction [1]. According to the phase diagram of Ag-Sn binary alloy [2], Sn is more soluble in ductile Ag-fcc phase (up to 11.5 at%) than in ductile Cu-fcc phase (up to 9.1 at%). Recently we found that the Ag-Sn $\zeta$ phase including Sn from 11.8 to 22.85 at% is also ductile at room temperature. Therefore we investigated the Nb$_3$Sn formation through diffusion reaction between Ag-Sn alloy and Nb by making the single-core Nb/Ag-Sn composite wires and multifilamentary Nb/Ag-Sn composite wires. Nb$_3$Sn is formed through the diffusion reaction between Nb and Ag-Sn alloy, and shows $T_c$ of 15-17.7 K and $B_{c2}(4.2 \text{ K})$ of 16-20 T. However, $I_c$ of the single-core wire is not so high, because the thickness of Nb$_3$Sn layer is very thin. The formation rate of Nb$_3$Sn between Nb/Ag-Sn is 1/10-1/100 times slower than those between Nb/Cu-Sn. However the $J_c$ of the Nb$_3$Sn layer between Nb/Ag-Sn is relatively high and comparable to that of the bronze processed Nb$_3$Sn layer without Ti or Ta additions [3]. Therefore the multifilamentary Nb$_3$Sn wire made by Nb/Ag-Sn micro-composite is interesting as a practical superconductor. Ag is apparently not as effective in accelerating the Nb$_3$Sn formation rate in the diffusion reaction as Cu.
2. Sample preparations and measuring methods

Ag-9at%Sn (fcc phase), Ag-12at%Sn (ζ phase), and Ag-24at%Sn (ε phase) alloy ingots were fabricated by using a Tammann furnace. The Ag-9at%Sn and Ag-12at%Sn alloys are ductile, and cold-worked into rods, while the Ag-24at%Sn alloy is brittle, and crashed into powder. The Ag-9at%Sn and Ag-12at%Sn rods were inserted into Nb pipes. The Ag-24at%Sn powder was also packed into a Nb pipe. These single-core composites were cold-drawn into fine wires with 0.79 mm diameter.

200 short single-core wires with 1.05 mm diameter were bundled and inserted into Nb pipes. The 200-core composite wires were cold-drawn into fine wires with 0.79 mm diameter, as shown in Figure 1. The same processes were repeated for the 200-core composite wires with Ag-9at%Sn in order to fabricate the 40000-core composite wires with Ag-9at%Sn. The single-core composite wire with the Ag-24at%Sn powder showed abnormal deformation in the cross-section configuration. Therefore we did not try to fabricate multi-core composite wire by using the Ag-24at%Sn powder.

Finally these single, 200, and 40000-core composite wires with 0.79 mm were heat treated at 600-1000°C to form Nb₃Sn layer with A15 crystal structure through the diffusion reaction. After the heat treatment, the Nb₃Sn wires were electroplated with Cu of 7 μm thick in order to solder the current and potential leads for $T_c$ and $I_c$ measurements.

$T_c$ was measured by a resistive method with a transport current of 0.01 A, and defined as the temperature where the specimen showed half the value of normal-state resistance. $I_c$ was defined as the transport current where the sample showed a potential rise of 100 μV/m with increasing the transport current in perpendicular fields. $B_{c2}$ was determined by extrapolation from Kramer plot by using $I_c$ in high fields [4]. OM and SEM observations were performed on the cross-sections of these wires in order to clear up the diffusion reactions between Nb and Ag-Sn alloys. The composition analysis at the diffusion layers were carried out with energy dispersive X-ray (EDX) spectrometer.

3. Results and discussions

According to the OM and SEM observations on the cross-section of the single-core wire, abnormal deformations in the cross-section configuration were occurred in the single-core wire with Ag-24at%Sn powder, while those were not observed in the single-core wires with Ag-9at%Sn and Ag-12at%Sn. Figure 1 shows the cross-sections of 200-core composite wires with Ag-9at%Sn and Ag-12at%Sn. Both the wires show the clear multifilamentary structures without any contacts between filaments. The Ag-12at%Sn cores in the figure is much smaller than the Ag-9at%Sn cores, because the Nb pipe with thicker wall was used for fabricating single-core Nb/Ag-12at%Sn composite.

Through SEM observations, we could observe a very thin layer (about 0.4 μm) is formed between Nb and Ag-Sn alloy, as shown in Figure 2. By considering the results of superconducting properties and EDX analysis on the thin layer between Nb and Ag-Sn alloy should be Nb₃Sn. Ag does not accelerate the Nb₃Sn formation in the diffusion reaction between Nb and Sn, as Cu does. The formation rate of Nb₃Sn between Nb/Ag-Sn is 1/10-1/100 times slower than those between Nb/Cu-Sn.
With the heat treatment at 650-900°C, the composite wires showed $T_c$ of 15-17.8 K and $B_{c2}(4.2$ K) of 15-20 T, which indicate that the formation of Nb$_3$Sn occurred with the diffusion reaction. Dependence of $T_c$, $I_c$(4.2 K, 14 T), and $B_{c2}(4.2$ K) on the Sn content in the Ag-Sn core are shown in Figure 3. The maximum values are used in this figure. In addition $I_c$ values were revised in this figure by using the total Nb/Ag-Sn boundary-lengths, in order to have the same boundary lengths as those in the Nb/Ag-9at%Sn composite wires.

With increasing Sn content in Ag-Sn core, $T_c$, $B_{c2}$, and $I_c$ are monotonically increased. With increasing core number, which indicates the decreasing of diffusion reaction length, $T_c$, $B_{c2}$, and $I_c$ are increased, excepting for $T_c$ degradation of 40000-core Nb/Ag-9at% Sn wire. The shorter length in diffusion reaction may cause an enough supply of Sn into the Nb$_3$Sn layer, resulting in the fabrication of near-stoichiometry Nb$_3$Sn with high $T_c$ and high $B_{c2}$. By the way, the core thicknesses in 40000-core Nb/Ag-9at% wire are less than 0.3 μm. The proximity effect may cause the $T_c$ degradation [5].

Figure 2. SEM micrograph on the boundary between Nb and Ag-9at%Sn in the 200-core wire heat treated at 800°C for 40 hr. The results of EDX analysis are also shown at lower side. The line analysis was performed at the position shown by a white line.

Typical $I_c$ vs. B curves for single-core, 200-core, and 40000-core Nb/Ag-9at%Sn composite wires are shown in Figure 4. With the increase of core number from 1 to 40000, $I_c$ of the wires are increased by 100 to 500 times. Of course the best $I_c$ value, obtained in this study, is also not enough for the practical applications. However, the $J_c$(4.2 K, 17 T) values of Nb$_3$Sn layers are higher than...
100 A/mm² for the 200-core, according to the SEM observations. In addition we could not measure the thickness of Nb₃Sn layer in the wire, heat treated for shorter time, with showing relatively high $I_c$, in which much higher $J_c$ can be expected. Therefore $J_c$ of the Nb₃Sn layer between Nb/Ag-Sn is comparable to that of the bronze processed Nb₃Sn conductor without Ti or Ta additions. By optimizing the cross-sectional configuration, the multifilamentary Nb₃Sn conductor made by Nb/Ag-Sn micro-composite should show higher overall $J_c$ in high fields.

The 40000-core wire with Ag-12at% Sn is under fabrication, of which results will be reported in near future.

4. Conclusions
Through these studies, we obtained the following conclusions.

(1) Nb₃Sn is formed through the diffusion reaction between Nb and Ag-Sn alloy.

(2) With increase of Sn content in Ag-Sn alloy, Nb₃Sn shows higher superconducting properties.

(3) Ag-Sn $\zeta$ phase shows enough ductility to fabricate a multifilamentary Nb/Ag-Sn composite wire.

(4) The formation rate of Nb₃Sn between Nb/Ag-Sn is 1/10-1/100 times slower than those between Nb/Cu-Sn.

(5) $I_c$ and $B_{c2}$, obtained for Nb/Ag-Sn conductor are similar to those for Nb/Cu-Sn conductor without Ti or Ta additions.

(6) $I_c$ is relatively small for single-core wire, and improved drastically by increasing core number, because $J_c$ of Nb₃Sn layer is relatively high. Therefore the Nb₃Sn conductor made by the new process seems to be practically interesting.

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