Large and field-insensitive critical current densities in (Sr,Na)Fe$_2$As$_2$ superconducting tapes

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Abstract. We have fabricated (Sr,Na)Fe$_2$As$_2$ tapes by optimizing the sintering temperature and time, and characterized their transport $J_c$ at 4.2 K up to 14 T. The transport $J_c$ for the (Sr,Na)Fe$_2$As$_2$ tape sintered at 875°C reaches $4.9 \times 10^4$ A/cm$^2$ under self-field, and it is suppressed only down to $1.9 \times 10^4$ A/cm$^2$ even at 14 T. We succeeded in fabricating very field-insensitive tapes, which are very promising for practical applications. We also report compositional distributions and magneto-optical images of current distribution in tapes. Magneto-optical imaging of the core of the tape clarified inhomogeneous intergranular current distributions.

1. Introduction

The iron-based superconductor was first discovered by the Hosono group in 2008 [1]. Soon after the discovery, several types of high temperature superconductivity in iron pnictides were discovered. Among them, superconducting wires and tapes have been fabricated mainly by using (AE,K)Fe$_2$As$_2$ materials (AE = Ba or Sr), so called ‘122-type’ [2-6], because of their high upper critical field $H_{c2}(0)$ [7, 8] and low anisotropy $\gamma$ (~2) [8, 9], as well as their high transition temperature $T_c$ (~36-38 K) [10, 11]. At present, the transport $J_c$ for these tapes has reached $10^5$ A/cm$^2$ [12, 13], which is the level for practical applications, at 4.2 K under high magnetic fields.

In addition to these K-doped materials, there are analogous Na-doped ones: (AE’,Na)Fe$_2$As$_2$ (AE’ = Ba, Sr, or Ca). Among them, polycrystalline samples of (Sr$_{1-x}$Na$_x$)Fe$_2$As$_2$ with the maximum $T_c$ of 36.5 K for $x = 0.55$ [14], and large transport $J_c$ at 20 K in a (Sr,Na)Fe$_2$As$_2$ tape were reported [15]. Although these Na-doped materials are very promising, superconducting wires and tapes using Na-doped ones have not been studied extensively.

In this paper, we report the relationship between transport $J_c$ of (Sr,Na)Fe$_2$As$_2$ tape and sintering temperature. The value of transport $J_c$ was the largest when the tape was sintered at 875°C, although the core of tape was reacted with silver sheath. In the tape which was sintered at 850°C for 2.5 h, the value of $J_c$ was almost as high as that of the tape which was sintered at 875°C, and the surface of silver sheath has not been reacted with the core. We performed a scanning electron microscope observations, compositional analyses with an energy dispersive x-ray spectroscopy (SEM-EDX), and magneto-optical (MO) observations for the tape sintered at 850°C.
2. Experiment
In fabricating superconducting tapes of (Sr$_{1-x}$Na$_x$)Fe$_2$As$_2$, we basically followed the method reported in Ref. [15]. First, we synthesized polycrystalline samples of (Sr$_{1-x}$Na$_x$)Fe$_2$As$_2$. Since it is reported that a sample with Na concentration $x = 0.55$ exhibited the highest $T_c$ (36.5 K), we selected $x$ as 0.55 [14]. Polycrystalline samples were synthesized at 720°C for 18 h using the stainless steel pipe, where pellets of starting materials (SrAs, NaAs, Fe$_2$As) were placed. Then, the reacted materials were ground into powder, pelletized, and reacted at 720°C 18 h again. The prepared polycrystalline materials were ground into powder with an agate mortar in a nitrogen filled glove box. The ground powder was tightly packed into a silver tube with outer and inner diameters of 4.6 and 3 mm, respectively. The tube was groove-rolled into a wire with 1.3 x 1.3 mm$^2$ cross section. Then, the wire was flat-rolled into a tape form with a thickness and width of 0.25 and 2.2 mm, respectively. The tape was cold-pressed under a pressure of 0.54 GPa. Finally, the tape was heated for sintering. Sintering temperature was 800°C, 825°C, 850°C, 860°C, and 875°C, and the sintering temperature dependence of $J_c$ was studied.

The $J_c$ was measured at 4.2 K by the standard four-probe method in magnetic fields up to 14 T. Powder x-ray diffraction (XRD) measurements were conducted using CuKα radiation. Magnetic measurements were conducted using a commercial SQUID magnetometer (MPMS-XL5, Quantum Design). Compositional analyses and visualization of magnetic induction in the core were conducted using SEM-EDX (S-4300, Hitachi High-Technologies equipped with EMAX x-act, HORIBA) and MO imaging, respectively.

3. Result and discussion
3.1. Transport $J_c$ measurements at 4.2 K in magnetic fields up to 14 T
Figure 1(a) shows $J_c$ versus magnetic field at 4.2 K for the (Sr,Na)Fe$_2$As$_2$ tapes sintered at various temperatures. The $J_c$ in (Sr,Na)Fe$_2$As$_2$ tape sintered at 875°C reaches 4.9 x 10$^4$ A/cm$^2$ under self field, and it is suppressed only down to 1.9 x 10$^4$ A/cm$^2$ even at 14 T. Although $J_c$ in 122 type superconducting tapes usually goes down most sharply in the range between 0 T and 2 T [16], $J_c$ in our tapes sintered at 875°C was hardly suppressed in this range. $J_c$(14 T) / $J_c$(0 T) was calculated from figure 1(a), and plotted in figure 1(b) for each sintering temperature. For the tape sintered at 875°C, $J_c$(14 T) / $J_c$(0 T) = 0.39. For (Ba,K)Fe$_2$As$_2$ tape, $J_c$(0 T) = 2.5 x 10$^4$ A/cm$^2$, and $J_c$(14 T) = 1.9 x 10$^4$ A/cm$^2$, then $J_c$(14 T) / $J_c$(0 T) = 0.08 [16]. For Nb$_3$Sn tape, $J_c$(0 T) = 8.2 x 10$^5$ A/cm$^2$, and $J_c$(14 T) = 1.0 x 10$^5$ A/cm$^2$, then $J_c$(14 T) / $J_c$(0 T) = 0.13 [17]. This indicates that suppression of $J_c$ by magnetic field in (Sr,Na)Fe$_2$As$_2$ tape is very weak.

From figures 1(a) and (b), it is suggested that the tapes sintered at higher temperature exhibit higher and more field-insensitive value of $J_c$. However, if the tape is sintered at temperatures over 875°C, severe reaction between the core and Ag sheath was observed. So, a range between 850 and 875°C is a critical region where the core begins to melt and reacts with the silver sheath. Probably, this partial melting of the core leads to improvement of connections between grains and higher inter-granular $J_c$.

3.2. X-ray diffraction analyses
Figure 2 shows XRD patterns for the core surface of the (Sr,Na)Fe$_2$As$_2$ tapes sintered at various temperatures. Compared with the reference peak of polycrystalline samples, the intensity of (002) peak is higher. This result indicates that the core is well textured with its c-axis perpendicular to the tape plane. We believe that this is due to the uniaxial cold press and it helps to achieve higher value of $J_c$. Also, we can identify the FeAs impurity peaks in tapes sintered at 800°C and 825°C, while this peak vanishes in the tape sintered at 850°C. The presence of impurity peaks at 800°C and 825°C partially explains why $J_c$ of the tapes sintered at temperatures over 850°C is higher. In addition, we can identify the Fe$_2$As impurity peaks in all tapes. Fe$_2$As may have been produced by the reaction between
Figure 1. (a) $J_c$ versus magnetic field at 4.2 K for the (Sr,Na)Fe$_2$As$_2$ tapes sintered at various temperatures. (b) $J_c(14 \text{T}) / J_c(0 \text{T})$ as a function of sintering temperature calculated from the data in figure 1(a). For the tape sintered at 875°C, $J_c(14 \text{T}) / J_c(0 \text{T}) = 0.39$. This means that critical current densities are very field-insensitive.

Ag sheath and As as will be shown in section 3.3, which decreased the amount of As in the core and helped the formation of Fe$_2$As. Alternatively, when the tape was sintered, As in the core may have vaporized and escaped from the core. Whichever the case, addition of As to the starting powders may reduce the Fe$_2$As impurity phase in the core, and contribute to enhance the transport $J_c$.

Figure 2. XRD patterns for the core surface of the (Sr,Na)Fe$_2$As$_2$ tapes sintered at various temperatures.

3.3. SEM images and compositional analyses
Figures 3(a) and (b) show SEM images of the surface of the core of the tape sintered at 850°C. Atomic ratios in four different regions on the surface of the tape marked in figures 3(a) and (b) are
The ideal atomic ratio is Sr : Na : Fe : As = 9 : 11 : 40 : 40 for x = 0.55. In region 1, there are impurities which seem like Fe$_2$As. In region 2, compounds of Fe and As and Ag are identified. This is consistent with SEM images reported in Ref. [15]. According to their report, there is a possibility that Ag sheath reacts with the core and penetrates into the core, and that these compounds of Fe and As and Ag partially complement impurity regions. These compounds are identified only in tapes sintered at temperatures over 850°C and can contribute to the large and field-insensitive $J_c$. In regions 3 and 4, atomic ratios are approximately equal to those of Sr$_{0.45}$Na$_{0.55}$Fe$_2$As$_2$, consistent with the starting composition.

| atom | Region 1 ratio (%) | 2 ratio (%) | 3 ratio (%) | 4 ratio (%) |
|------|---------------------|-------------|-------------|-------------|
| Sr   | 4.2                 | 2.7         | 10.1        | 7           |
| Na   | 5.9                 | 4.5         | 12.4        | 11          |
| Fe   | 50.7                | 12.8        | 37.1        | 44          |
| As   | 39.2                | 12.5        | 40.1        | 37.3        |
| Ag   | 0.1                 | 67.5        | 0.3         | 0.7         |

3.4. MO imaging

Figure 4(a) shows MO images of the surface of the tape sintered at 850°C. This image was taken at 5 K after cycling the magnetic field up to 0.1 T and back to zero. Bright regions trap higher magnetic field, and correspond to grains that are well connected. Figure 4(b) shows temperature dependence of residual magnetic induction along the red line in figure 4(a). Thickness of the core is roughly 100 μm, and the highest trapped field is approximately 60 mT. From this trapped field value, we can roughly evaluate $J_c \sim 6 \times 10^4$ A/cm$^2$. Figures 4(c) and (d) show optical images of the surface of the tape sintered at 850°C. In figure 4(c), the bright region on the left side shows an unpolished Ag sheath. In figures 4(c) and (d), slightly bright spots are high-density regions of (Sr,Na)Fe$_2$As$_2$ grains corresponding to bright regions in figure 4(a). These images indicate that high-density regions are localized, and that there are some regions where grains are weakly connected each other and intergranular $J_c$ is small. To improve the connectivity between grains, the hard and homogeneous core in the tape should be realized, and this will lead to further enhancement of $J_c$. 

Figure 3. (a) (b) SEM images of the surface of the tape sintered at 850°C.

Table 1. Atomic ratios in four different regions on the surface of the tape marked in figures 3(a) and (b).
Figure 4. (a) MO images of the surface of the tape sintered at 850°C. Bright regions are trapping higher magnetic field. (b) Residual magnetic induction profile along the red line in (a). (c)-(d) Optical images of the surface of the tape sintered at 850°C. In (c), bright region on the left side shows an unpolished Ag sheath. In (c) and (d), slightly bright spots are high-density regions of (Sr,Na)Fe$_2$As$_2$ grains and correspond to bright regions in (a).

4. Summary
We have reported the fabrication and characterization of (Sr,Na)Fe$_2$As$_2$ superconducting tapes prepared under different conditions including transport $J_c$ up to 14 T and magneto-optical observation of the trapped field. In the tape sintered at 875°C, the value of $J_c$ reached 4.9 x 10$^4$ A/cm$^2$ under self-field, and it is suppressed only down to 1.9 x 10$^4$ A/cm$^2$ even at 14 T. We succeeded in fabricating very field-insensitive tapes, which are very promising for practical applications. Magneto-optical imaging of the core of the tape clarified inhomogeneous intergranular current distributions.

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