FeSe superconducting tapes with a high critical current density fabricated by diffusion method

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Abstract. We have prepared high-quality FeSe tape through diffusion method. The transport critical current density evaluated by the current-voltage characteristics is 600 A/cm² at 4.2 K under zero field. The upper critical field of the FeSe tape at T = 0 K estimated by Werthamer-Helfand-Hohenberg theory is 243 kOe, which is similar to that of single crystals.

1. Introduction
Since the discovery of iron-pnictide LaFeAsO¹ₓFₓ showing superconductivity ~ 26 K [1], intensive efforts have been devoted to the iron-based superconductors. Among these studies, a binary superconducting compound FeSe with the simplest structure has been discovered soon [2]. Research directed to the applications has been under way because of its high upper critical field [3]. Powder-in-tube (PIT) method is the most common method for making superconducting wires in iron-based superconductor system, and also being adopted in fabricating FeSe wires [4]. In PIT method, rolling and drawing processes are needed to make the raw materials denser. Diffusion method was originally adapted to fabrication of A15 compounds, such as Nb₃Sn and V₃Ga [5]. It usually proceeds in a composite, composed of a high melting point component and a low melting point component, which results in the formation of new phases stable at the reaction temperature. The high-Tc superconducting cuprates and MgB₂ wire and tapes have also been fabricated by the diffusion method in a shorter reaction time than by a conventional sintering process [6, 7]. Recently, fabrications of FeSe wires and tapes using diffusion method have been reported [4, 8]. Here we report fabrication of high-quality FeSe tapes through diffusion method. The characterization through X-ray diffraction, magnetization, resistivity, transport critical current density, and magneto-optical measurements are discussed.

2. Experiments
Se grains and iron tapes were used as raw materials. Iron tapes are cut into dimensions ~ 50×5×0.3 mm³. Proper amounts of Se grains and iron tapes were sealed in an evacuated quartz tube. The sealed tapes and grains were put into a furnace and heated up to 800 °C with a sweeping rate of 100 °C/h, and kept at this temperature for 12 h. Then the furnace was switched off and cooled to room temperature naturally. The structure of the sample was characterized by using X-ray diffraction (M18XHF, MAC Science) with Cu-Kα radiation generated at 40kV and 200 mA. Bulk magnetization is measured by a superconducting quantum interference device SQUID magnetometer (MPMS-5XL, Quantum Design). Microstructures were characterized by Scanning Electron Microscope (SEM, Hitachi S-4300) operated

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at 15 kV. Resistivity and current-voltage (I-V) measurements were performed by the four-probe method. Resistivity measurements were performed within the sample chamber of a SQUID magnetometer. I-V measurements were performed by immersing the tape into liquid helium. For local magnetic characterization, magneto-optical (MO) imaging was applied. A Bi-substituted iron-garnet indicator film is placed in direct contact with the sample, and the whole assembly is attached to the cold finger of a He-flow cryostat (Microstat-HR, Oxford Instruments) and cooled down to 5 K. MO images are acquired by using a cooled CCD camera with 12-bit resolution (ORCA-ER, Hamamatsu). To enhance the visibility of the local magnetic induction and eliminate the signals from the impurity phases, a differential imaging technique is employed [9, 10].

3. Results and discussion

As-prepared FeSe tape is shown in Figure 1(a). Figure 1(b) shows the X-ray diffraction pattern of as-prepared tape. All the peaks are well indexed using a space group of P4/nmm. The compound crystallizes in a tetragonal structured FeSe. The calculated lattice constants are $a = 0.3759$ nm, and $c = 0.5505$ nm. The lattice parameter $c$ is slightly smaller than that in FeSe single crystals [3]. Temperature dependences of zero-field-cooled (ZFC) and field-cooled (FC) magnetization at 5 Oe of the FeSe tape are shown in Figure 1(c). The sample shows an onset of diamagnetism at around 8 K. Inset of Figure 1(c) shows magnetic hysteresis curves of this sample. From the magnetization hysteresis loops, we can evaluate intragranular critical current density $J_{c,\text{intra}}$ for polycrystalline samples using the Bean model with an assumption of field-independent $J_c$. According to the Bean model, $J_{c,\text{intra}}$ [A/cm$^2$] is given by

$$J_{c,\text{intra}} = \frac{30 \Delta M}{d},$$

with an assumption that intergranular critical current is zero, where $\Delta M$[emu/cc] is $M_{\text{up}} - M_{\text{down}}$, $M_{\text{up}}$ and $M_{\text{down}}$ are the magnetization when sweeping field up and down, respectively, $d$[cm] is the average diameter of the grain in the polycrystalline sample [11]. $J_c$ calculated from $M$-$H$ curve is estimated to be $1 \times 10^5$ A/cm$^2$ at 5 K under zero field.

![Figure 1](image_url)

**Figure 1.** (a) Photograph of as-prepared FeSe tape. (b) X-ray diffraction pattern of FeSe tape. (c) Temperature dependence of magnetization of FeSe tape measured at 5 Oe. Inset: Magnetic field dependence of magnetization at 2 K and 5 K.

Figure 2(a) is the SEM image of a part of this FeSe tape separated from the iron core. The sample dimensions are $875 \times 775 \times 50$ μm$^3$. From this image, the typical grain size is ~ 20 μm. Figures 2(b) and 2(c) depict MO images of the FeSe tape in the remanent state after applying a 500 Oe field for 0.25 seconds which was subsequently reduced down to zero at 5 K and 8 K. In these figures, the bright regions correspond to the trapped flux in the sample. At both temperatures, the field profile is inhomogeneous, which implies that the intergranular current density is much smaller compared with the intragranular current density. The intragranular current density decreases gradually as the temperature is increased towards $T_c$. We calculated the intragranular critical current density from the magnetic induction profile. Figure 2(d) shows the magnetic induction profiles along the dotted line in
Figure 2(b). In this calculation, we roughly estimate the intragranular critical current densities by $J_c \sim dB/dx$. For typical grains, $J_c$ thus estimated is $\sim 1 \times 10^4$ A/cm$^2$ at 5 K. This value is much smaller than that estimated from the $M-H$ curve. The reason for this difference may be similar to the case of the FeTe$_{0.5}$Se$_{0.5}$ polycrystalline sample [12]. Figures 2(e) and 2(f) reveal the penetration of vortices at 5 K in the FeSe tape. In Figure 2(e), at low field of 1 Oe, most parts of the sample are still in the Meissner state. When the field is increased to 3 Oe, magnetic flux penetrates intergranular regions. The shapes of the superconducting grains are more visible when the field is increased. This information indicates the presence of weak links between superconducting grains.

We investigated the transport $J_c$ of this FeSe tape. The FeSe tape dimensions for this measurement are 50×5×0.4 mm$^3$, and the distance between two voltage contacts is 0.82 mm. Figure 3(a) shows the zero-field $E-J$ characteristics at 4.2 K. Here we adopt $E = 1 \mu$V/cm as a criterion for the $E-J$ curve to define transport $J_c$. At 4.2 K, transport $J_c$ as high as 600 A/cm$^2$ is observed in this FeSe tape. This is the highest value among single-core FeSe wire and tapes. Figure 3(b) shows the variation of $T_c$ with magnetic field for $H = 0, 10, 20, 30, 40$ and $50$ kOe. With increasing field, the resistive transition shifts to lower temperatures accompanied by a slight increase in the transition width. Inset of figure 3 shows the variation of the upper critical field $H_{c2}$ with reduced temperature $t = T/T_{c \text{onset}}$ for the FeSe tape. The values of $H_{c2}$ were defined as the field at the midpoint of the resistive transition. The slope of $H_{c2}$ at $T_c$ is -26.4 kOe K$^{-1}$. The value of $H_{c2}$ at $T = 0$ K estimated using the Werthamer–Helfand–Hohenberg formula [13], $H_{c2}(0) = -0.69T_c \partial H_{c2}/\partial T|_{T=T_c}$, is 243 kOe. This value is comparable to other reported FeSe wire and tapes [4, 8]. In order to extract the superconducting parameters, we have used the Ginzburg–Landau (GL) formula for the coherence length ($\xi$), $\xi = (\Phi_0/2\pi H_{c2})^{1/2}$, where $\Phi_0 = 2.07 \times 10^{-7}$ G cm$^2$, the coherence length $\xi$ at the zero temperature is calculated as 3.64 nm.

In superconducting wires and tapes, both $J_c$ and critical current ($I_c$) are important parameters for applications. Using our diffusion method, it is easy to increase $I_c$ by simply increasing the cross
section of the raw Fe tape. The quality and performance of FeSe tapes can be improved by changing the ramping rate and sintering temperature to control the vapor pressure, and the cooling process or introducing an annealing step.

**Figure 3.** (a) $E$–$J$ characteristics in FeSe tape at 4.2 K. (b) Field dependence of the resistivity of FeSe tape around $T_c$. Inset shows the upper critical field $H_{c2}$ versus temperature determined by the midpoint of the resistive transition.

4. Conclusion

In summary, X-ray diffraction, magnetization, resistivity, transport critical current density and magneto-optical measurements were performed on high quality FeSe tape fabricated by diffusion method. The transport critical current density 600 A/cm$^2$ at 4.2 K under zero field is obtained. This value is more than two orders less than the intragranular critical current density. The upper critical field $H_{c2}$ of the FeSe tape at $T = 0$ K is estimated as 243 kOe, which is comparable to that of single crystals. The diffusion method is promising for fabrication of iron-based superconducting tapes and wires.

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