Transport properties of iron-based FeTe$_{0.5}$Se$_{0.5}$ superconducting wire

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Abstract—FeTe$_{0.5}$Se$_{0.5}$ superconducting wires have been fabricated using ex-situ PIT method with an Fe sheath. Among the other FeAs-based superconductors, FeTe$_{0.5}$Se$_{0.5}$ has great advantage for applications due to its binary composition and less toxicity. Surprisingly, superconducting current was observed in the as-fabricated wire without any heat treatments. The zero-resistivity critical temperature ($T_c$) and transport critical current density ($J_c$) at 2 K were 3.2 K and 2.8 A/cm$^2$, respectively. In addition, by annealing at 200°C for 2 h, $T_c$ and $J_c$ were enhanced up to 9.1 K, and 64.1 A/cm$^2$, respectively. This suggests that the inter-grain connectivity was improved by heat treatment, and superconducting properties of FeTe$_{0.5}$Se$_{0.5}$ wire were enhanced.

Index Terms—FeTe$_{0.5}$Se$_{0.5}$; Superconducting wires; Critical temperature; Critical current density;

I. INTRODUCTION

The discovery of iron-based superconductors [1] inspired an immense research activity in this field. Over the past two years, several groups of iron-based superconductors have been discovered, such as BaFe$_2$As$_2$ (122 series) [2], LiFeAs (111 series) [3] and FeSe (11 series) [4]. Among them, The 11 series, such as FeSe, FeTe$_{1-x}$Se$_x$, and FeTe$_{1-x}$S$_x$, is an important ferrous superconducting system. The 11 series has the simplest structure without block layers along the c-axis, and less toxicity compared to the other FeAs-based superconductors [5]-[8]. Although the transition temperature ($T_c$) of FeSe is as low as 8.5 K, the $T_c$ can be highly enhanced to 15 K by partial substitution of Te or S [5]-[10], and up to 37 K under high pressure [11]-[13]. Moreover, 11 series have a high upper critical field ($H_{c2}$) [6], [14], [15]. Therefore, the 11 series have a great potential for applications.

We succeeded in the observation of a zero-resistivity current on the current-voltage measurement for the iron-based superconducting wire using the in-situ powder-in-tube (PIT) method with an Fe sheath [16]. Up to now, there are hardly any new reports on the fabrication of superconducting wires in which transport critical current density ($J_c$) values were obtained [16], [17]. Furthermore, obtained $J_c$ in these wires is much lower than intra-grain $J_c$ [18]. Lee et al. pointed out that grain boundaries in 122 series exhibit current-limiting behavior similar to that observed in high-$T_c$ cuprates [19]. However, heat treatment may enhance grain connectivity and result in $J_c$ improvement [20]. In this paper, we report on the fabrication of the FeTe$_{0.5}$Se$_{0.5}$ superconducting wire using the ex-situ PIT method with an Fe sheath, and the effect of heat treatment on superconductivity in FeTe$_{0.5}$Se$_{0.5}$ wire.

II. EXPERIMENTAL

FeTe$_{0.5}$Se$_{0.5}$ wires were prepared using an ex-situ PIT method with an Fe sheath. First of all, polycrystalline samples of FeTe$_{0.5}$Se$_{0.5}$ were prepared using the solid state reaction method. High purity powders of Fe (99.9% up), Se (99.999%) and Te (99.999%) were mixed with nominal compositions and sealed into evacuated quartz tubes. Then the powders were heated at 650°C for 15 h. The obtained samples were ground and pressed into pellets. The pellets were reheated at 650°C for 15 h in evacuated quartz tubes. After furnace cooling, the product was reground and packed into a pure Fe tube with a length of 5 cm. The inner and outer diameters of the tube were 3.5 and 6 mm, respectively. The composite was initially fabricated in a rod shape with about 2.5 mm × 2.5 mm cross section by groove rolling and then drawing into a wire of 1.1 mm in diameter. Fig. 1(a) shows optical micrographs of the transverse cross section of the wire. The cross section shows

![Fig. 1. FeTe$_{0.5}$Se$_{0.5}$ wire fabricated by the ex-situ PIT method. (a) Optical micrograph of the transverse cross section of wire after heat treatment at 200°C for 2 h. (b) Photograph of samples after cutting and before heating.](image-url)
uniform deformation of composite without any breakage. The round-shaped wire was cut into pieces of about 4 cm in length (Fig. 1(b)), and the wires were sealed into a quartz tube with an atmospheric-pressured argon gas. The sealed wires were rapidly heated at 150-500 °C for 2 h.

Constituent phases were determined by powder X-ray diffraction (XRD) with Cu Kα radiation and intensity data were collected in the 2θ range of 5-60°. Temperature dependence of magnetic susceptibility was measured using a superconducting quantum interference device (SQUID) magnetometer at H = 10 Oe. Temperature dependence of resistivity and transport critical current density \(J_c\) were measured by the four-probe method with a physical property measurement system (PPMS).

III. RESULT AND DISCUSSION

A. Polycrystalline \(\text{FeTe}_{0.5}\text{Se}_{0.5}\)

The x-ray diffraction pattern of \(\text{FeTe}_{0.5}\text{Se}_{0.5}\) compound is shown in Fig. 2. All the peaks were well indexed using a space group of P4/mnm except for an unidentified small peak as an impurity phase labeled by asterisks. The compound crystallizes in a tetragonal structure and no secondary phase is observed. Lattice parameters are found to be \(a=3.7943(2)\) Å and \(c=5.9966(5)\) Å. The lattice parameters are in good agreement with previously published reports [5], [21].

Temperature dependence of zero-field-cooled (ZFC) and field-cooled (FC) magnetization of polycrystalline \(\text{FeTe}_{0.5}\text{Se}_{0.5}\) is shown in Fig. 3(a). The sample showed superconducting transition at 13.9 K. The superconducting volume fraction estimated from the ZFC magnetization at 2 K was larger than 100% due to the porous structure and a demagnetization effect of the sample. Fig. 3(b) shows the temperature dependence of the resistivity of \(\text{FeTe}_{0.5}\text{Se}_{0.5}\) compound. Resistivity around \(T_c\) is shown in the inset. While this compound has a somewhat semiconducting behavior above 150 K, it shows metallic behavior in the normal state below 150 K. Similar temperature dependence of resistivity is reported for samples with compositions close to \(\text{FeTe}_{0.5}\text{Se}_{0.5}\) [7], [8], [21], [22]. This behavior has been attributed to a weak charge-carrier localization due to a large amount of excess Fe in \(\text{Fe}_{1+y}\text{Te}_{1-x}\text{Se}_x\) system [23]. Additionally, the value of resistivity is higher than that reported in the previous reports [7], [8], [21], [22]. This could be resulted from the microscopic inhomogeneities that are still present in our sample and impurity phases. The superconducting transition was observed at \(T_c^{\text{onset}}\) of 15.7 K and zero-resistivity was achieved at 12.8 K. These values are almost the same as previous reports on polycrystalline and single crystal \(\text{FeTe}_{0.5}\text{Se}_{0.5}\) samples [19]-[23].

B. \(\text{FeTe}_{0.5}\text{Se}_{0.5}\) superconducting wire

Fig. 4 shows the temperature dependence of resistivity at zero magnetic fields for \(\text{FeTe}_{0.5}\text{Se}_{0.5}\) wires fabricated by ex-situ PIT method. The values of \(T_c^{\text{onset}}\), \(T_c^{\text{zero}}\), and transition width (\(\Delta T_c\)) are listed in table 1. Surprisingly, superconducting current was observed in the as-fabricated wire without any heat treatments. The zero-resistivity critical temperature \(T_c^{\text{zero}}\) and transport critical current density \(J_c\) at 2 K were 3.2 K and 2.8 A/cm², respectively. \(J_c\) was obtained using the criteria of \(E = 1\) \(\mu\)V/cm. In addition, with increasing temperature of heat treatment, the value of \(T_c^{\text{onset}}\) and \(T_c^{\text{zero}}\) increased. The highest \(T_c^{\text{onset}}\), \(T_c^{\text{zero}}\) and \(\Delta T_c\) were 10.8 K, 9.1 K and 1.7 K for
reported that the superconducting transition in single crystal of superconducting phase of Fe (Se,Te) \[16\]. Moreover, it is Fe sheath reasonably supplied Fe for synthesizing the in-situ PIT method \[16\]. It is considered that the heat treatment were reduced. Furthermore, FeTe in FeTe \[16\] showed 82.5 $\mu$A/cm$^2$ at 2 K and 64.1 $\mu$A/cm$^2$ at 4.2 K, a factor of 5 higher than the value obtained Fe (Se,Te) wire fabricated by in-situ PIT method \[16\]. It is considered that the heat treatment in FeTe,0.5Se,0.5 improved the inter-grain connectivity. However, with an increase in temperature above 250$^\circ$C, the values of $T_c$ zero were reduced. Furthermore, FeTe,0.5Se,0.5 wires annealed above 400$^\circ$C did not show zero-resistivity. We recently confirmed that Fe sheath reasonably supplied Fe for synthesizing the superconducting phase of Fe (Se,Te) \[16\]. Moreover, it is reported that the superconducting transition in single crystal of Fe$_{0.5}$Te$_{0.5}$, suppressed by excess Fe \[23\]. Given these results, Fe sheath might excessively supply Fe for the superconducting phase, and the increase of excess Fe concentration resulted in reduced superconductivity.

Furthermore, it is notable that the $T_c$ onset values of each FeTe$_{0.5}$Se$_{0.5}$ wire were suppressed more than 4 K compared to than that of FeTe$_{0.5}$Se$_{0.5}$ polycrystalline bulk. This result could be attributed to stress of FeTe$_{0.5}$Se$_{0.5}$ phase due to rolling process.

Fig. 5(a) shows the temperature dependence of the resistivity under various magnetic fields for the FeTe$_{0.5}$Se$_{0.5}$ wire annealed at 200$^\circ$C for 2 h. With increasing magnetic fields, the $R(T)$ curves are shifted to lower temperature, but also they do not much broaden, indicating that a two-dimensional feature and the superconducting fluctuations in this wire are small. The upper critical field ($\mu_0H_{c2}$) and the irreversibility field ($\mu_0H_{irr}$) of the FeTe$_{0.5}$Se$_{0.5}$ wires annealed at 200$^\circ$C for 2 h are plotted in Fig. 5(b) as a function of temperature. We have estimated $\mu_0H_{c2}$ and $\mu_0H_{irr}$, using 90% and 10% of normal state resistivity, respectively. The $\mu_0H_{c2}(0)$ is estimated to be ~40 T by linear extrapolation. Applying the Werthamer-Helfand-Hohenberg (WHH) theory \[24\], the $\mu_0H_{c2}(0)$ is calculated to be ~28 T. In order to approximate the superconducting parameters, we have used the Ginzburg-Landau formula for the coherence length ($\xi$), $\xi = (\phi_0/2\pi\mu_0H_{c2})^{1/2}$, where $\phi_0 = 2.07$ Oe cm$^2$, $\xi(0)$ was calculated as ~3.4 nm. Additionally, as shown in Fig. 5(b), $\mu_0H_{irr}$ values at 4.2 K and 0 K are estimated to be ~13 T and ~25 T, respectively, by linear extrapolation. In polycrystalline FeTe$_{0.5}$Se$_{0.5}$, the $\mu_0H_{c2}(0)$ ~120 T was reported \[14\]. We found that the heat treatment improved superconducting properties due to better connections between grains. Superconducting properties can be further enhanced by improvement of inter-grain connectivity. Therefore, FeTe$_{0.5}$Se$_{0.5}$ wires have a
great potential for enhanced superconducting properties by optimization of the fabrication process.

IV. CONCLUSION

We fabricated FeTe$_{0.5}$Se$_{0.5}$ superconducting wire using the ex-situ PIT method with an Fe sheath. In the as-grown FeTe$_{0.5}$Se$_{0.5}$ wire without heat treatment, zero-resistivity was observed at 3.2 K and transport $J_c$ showed 2.8 A/cm$^2$ at 2 K. In addition, by annealing at 200°C for 2 h, $T_c$ zero and $J_c$ were enhanced up to 9.1 K and 82.5 A/cm$^2$ at 2 K, respectively. The heat treatment improved the superconducting properties such as $T_c$ and $J_c$ due to improvement of the inter-grain connectivity. For applications, it is desirable to improve the transport $J_c$ and $\mu_0H_{c2}$. FeTe$_{0.5}$Se$_{0.5}$ has high intra-grain $J_c$ and high $\mu_0H_{c2}$. Therefore, it is quite possible that significant improvement of transport $J_c$ and $\mu_0H_{c2}$ can be achieved by the enhancement of inter-grain $J_c$.

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