RAPID COMMUNICATION

Superconducting properties of granular SmFeAsO$_{1-x}$F$_x$ wires with $T_c = 52$ K prepared by the powder-in-tube method

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Abstract

We report the fabrication of Ta-sheathed superconducting SmFeAsO$_{1-x}$F$_x$ wires by the powder-in-tube method for the first time. The transition temperature of the SmFeAsO$_{0.65}$F$_{0.35}$ wires was confirmed to be as high as 52 K. High critical fields $H_{c2}(0) \geq 120$ T as well as current density $J_c$ of 3900 A cm$^{-2}$ at 5 K were also demonstrated. It should be noted that the $J_c$ exhibits a very weak field dependence behavior, indicating a very encouraging prospect for application of the new superconductors.

(Some figures in this article are in colour only in the electronic version)

The recent discovery of the iron-based superconductor LaFeAs(O$_{1-x}$F$_x$) with critical temperature 26 K has triggered great interest in this new family of high temperature superconductors [1]. Immediately, the $T_c$ has been enhanced to above 50 K in ReOFeAs (Re, rare-earth metal) by the substitution of La using other smaller rare-earth elements [2–7]. The present research indicates that the layered structure with the conducting Fe$_2$As$_2$ layers should be responsible for high temperature superconductivity, and the Re$_2$O$_2$ (Re = rare earth) layers behave as the charge reservoirs; all these look very similar to the case of cuprates. On the other hand, several groups have studied the $H_{c2}$, the irreversible magnetization and grain connectivity [8–15]. The resistive transition in fields suggests that $H_{c2}$ is remarkably high, which indicates encouraging potential applications. However, it is really a challenge to fabricate wires of ReOFeAs, because these materials are mechanically hard and brittle and therefore not easy to draw into the desired wire geometry. On the other hand, the use of proper metal cladding is another critical issue because of the strong chemical reactivity of ReOFeAs at high annealing temperatures of ~1200 °C. Recently, we have reported that Fe-sheathed LaFeAsO$_{0.95}$F$_{0.05}$ wires with Ti as a buffer were fabricated by the powder-in-tube (PIT) method [16]. In this work, we have synthesized tantalum-clad SmFeAsO$_{1-x}$F$_x$ wires for the first time and investigated the superconducting properties of SmFeAsO$_{1-x}$F$_x$ samples.

The SmFeAsO$_{1-x}$F$_x$ wires were prepared by the in situ powder-in-tube (PIT) method using Sm, As, SmF$_3$, Fe and Fe$_2$O$_3$ as starting materials. The raw materials were thoroughly grounded by hand with a mortar and pestle. The mixed powder was filled into a Ta tube of 8 mm outside diameter and 1 mm wall thickness. It is worth noting that the grinding and packing processes were carried out in a glove box filled with high purity argon atmosphere. After packing, the tube was rotary swaged and then drawn to wires of 2.25 mm in diameter. The wires were cut into 4–6 cm pieces and sealed in an Fe tube. They were then annealed at 1160–1180 °C for 45 h. The high purity argon gas was allowed to flow into the furnace during the heat-treatment process to reduce the oxidation of the samples.

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The phase identification and crystal structure investigation were carried out using x-ray diffraction (XRD). Standard four-probe resistance and magnetic measurements were carried out using a physical property measurement system (PPMS). The microstructure was studied using scanning electron microscopy (SEM/EDX) after peeling away the Ta sheath.

Figure 1 shows the optical images for typical transverse and longitudinal cross-sections of SmFeAsO$_{1-x}$F$_x$ wires after heat treatment. A reaction layer with a thickness 10–30 μm was formed between the superconducting core and the Ta tube due to the very high annealing temperature.

Figure 2 presents the XRD patterns of SmFeAsO$_{1-x}$F$_x$ wires after peeling off the sheath materials. As can be seen, the sample consists of a main superconducting phase, but some impurity phases are also detected. Such impurity phases might be reduced by optimizing the heating process and stoichiometry ratio of starting materials. The lattice parameters are $a = 3.9310(5)$ Å, $c = 8.522(1)$ Å for the sample with $x = 0.35$, and $a = 3.9275(5)$ Å, $c = 8.498(2)$ Å for the sample with $x = 0.3$, which indicates a clear shrinkage of lattice parameters by increasing substitution of F$^-$ for O$_2^-$.

Figure 3 shows the temperature dependence of resistivity at zero magnetic field for the samples SmFeAsO$_{1-x}$F$_x$ wires after peeling away the Ta sheath. With decreasing temperature, the electrical resistivity of the sample with $x = 0.35$ decreases monotonically, and a rapid drop was observed starting at about 52 K, indicating the onset of superconductivity. The sample with $x = 0.30$ shows the same behavior as that of the sample with $x = 0.35$, except for a slightly low onset $T_c$. The residual resistivity ratio RRR $= \rho(300 \text{ K})/\rho(55 \text{ K})$ was 2.8 and 2.3 for the samples $x = 0.35$ and $x = 0.3$, respectively, indicating stronger impurity scattering than LaFeAsO$_{1-x}$F$_x$ [17].

Figure 4 shows the SEM images, illustrating the typical microstructure of the fractured core layers for SmFeAsO$_{0.7}$F$_{0.3}$ samples. It can be seen in figure 4(a) that SmFeAsO$_{0.7}$F$_{0.3}$ has a dense microstructure with few voids. EDX analysis also revealed that the sample possibly contained impurity phases such as SmAs and unreacted Fe or Fe$_2$O$_3$. The crystal structure of this kind of material is based on a stack of alternating SmO and FeAs layers. A layered structure can clearly be seen, as shown in figure 4(b), very similar to what has been observed in Bi-based cuprates.

Magnetization loops with a strong ferromagnetic background were observed in our samples, which are believed to be contributed by the unreacted Fe or Fe$_2$O$_3$ impurity.
Figure 4. (a) Low magnification and (b) high magnification SEM micrographs for the SmFeAsO₋ₓFₓ wires.

Figure 5. Magnetic field dependence of J_c at 5 K for a bar and powder of SmFeAsO₁₋ₓFₓ wires.

Figure 6. Temperature dependence of resistivity under magnetic fields up to 9 T for the SmFeAsO₀.₆₅F₀.₃₅ wires. The inset shows the temperature dependence of H_c² and H_irr determined from 90% and 10% points on the resistive transition curves.

phases [10]. We calculated the global J_c for our samples on the basis of $J_c = 20\Delta M/a(1 - a/3b)$, where $\Delta M$ is the height of the magnetization loop and $a$ and $b$ are the dimensions of the sample perpendicular to the magnetic field, $a < b$. The intragrain $J_c$ was also evaluated on the basis of $J_c = 30\Delta M/(R)$ through magnetic measurements after grinding the superconducting core into powder. $(R)$ is the average grain size, which is about 10 μm, measured by SEM. The $J_c$ field dependence is shown in figure 5. It can be seen that $J_c$ based on the sample size is much lower than that what should exist in individual grains. The $J_c$ value at 5 K for the SmFeAsO₀.₆₅F₀.₃₅ powder is $2 \times 10^5$ A cm⁻², with a very weak dependence on field, which shows that SmFeAsO₀.₆₅F₀.₃₅ has a fairly large pinning force in the grain. However, the global $J_c$ value of 3.9 $\times$ 10⁷ A cm⁻² at 5 K obtained for the Sm samples is significantly lower than that seen in random bulks of MgB₂, which generally attained $10^8$ A cm⁻² at 4.2 K [18]. The main reason may be ascribed to the second impurity phases or the presence of weak links between the grains. As supported by XRD and SEM/EDX results, major impurity phases such as SmAs, SmOF, and unreacted Fe or Fe₂O₃ were observed in our present samples. These macroscale impurity phases can significantly reduce the percolating current path and limit the global $J_c$. Although the whole-sample current densities are significantly lower than that in randomly grain-oriented bulk of MgB₂, the grain connectivity seems better than that of random polycrystalline cuprates [12]. Recently, we have measured the transport critical current of SmFeAsO₁₋ₓFₓ wires at 4.2 K, unfortunately, we have not yet obtained the transport current. This is not surprising since the porosities, multiphase composition and most importantly extensive interfacial reaction between Ta and SmFeAsO₁₋ₓFₓ core have significantly reduced the percolating current path. In order to get the transport $J_c$, these issues should be solved first. Work on improving the superconducting properties of SmFeAsO₁₋ₓFₓ wires is in progress.

Figure 6 shows the resistive superconducting transitions for the SmFeAsO₀.₆₅F₀.₃₅ sample under magnetic fields after peeling away the Ta sheath. The large magnetoresistance at the normal state may be caused by magnetic impurity. It is found that the onset transition temperature is not sensitive to the magnetic field, but the zero resistance point shifts more quickly to lower temperatures due to the weak links or flux flow. We tried to estimate the upper critical field ($H_c²$) and
irreversibility field \((H_{irr})\), using the 90% and 10% points on the resistive transition curves. The inset of figure 6 shows the temperature dependence of \(H_{c2}\) and \(H_{irr}\) with magnetic fields up to 9 T for the SmFeAsO\(_{0.65}\)F\(_{0.35}\) samples. It is clear that the \(H_{c2}(T)\) curve is very steep, with a slope of \(-dH_{c2}/dT\rvert_{T_c} = 2.74 \text{ T K}^{-1}\). For a type II superconductor in the dirty limit, \(H_{c2}(0)\) is given by the Werthamer–Helfand–Hohenberg formula \([19]\), \(H_{c2}(0) = 0.693 \times (dH_{c2}/dT) \times T_c\). Taking \(T_c = 51\ \text{K}\), we get \(H_{c2}(0) \approx 96.8\ \text{T}\). On adopting a criterion of 99% \(\rho(T)\) instead of 90% \(\rho(T)\), the \(H_{c2}(0)\) value of this sample obtained by this equation is higher than 120 T. We can also see that the irreversibility field in the inset of figure 6 is rather high compared to that in MgB\(_2\). These high values of \(H_{c2}\) and \(H_{irr}\) indicate that this new superconductor has an encouraging prospect for application in very high fields.

We have prepared Ta-clad SmFeAsO\(_{1-x}\)F\(_x\) wires by the PIT process. The \(J_c\) of composite wires has a very weak dependence on magnetic field with a very high \(H_{c2}(0)\) above 120 T. Although some problems such as phase purity remain unsolved, our preliminary results explicitly demonstrate the feasibility of fabricating iron-based superconducting wires.

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