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Large transport critical currents of powder-in-tube $Sr_{0.6}K_{0.4}Fe_2As_2$/Ag superconducting wires and tapes

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We report the achievement of transport critical currents in $Sr_{0.6}K_{0.4}Fe_2As_2$ wires and tapes with a T_c = 34 K. The wires and tapes were fabricated through an in situ powder-in-tube process. Silver was used as a chemical addition as well as a sheath material. All the wire and tape samples have shown the ability to transport superconducting current. Critical current density J_c was enhanced upon silver addition, and at 4.2 K, a largest J_c of $\sim$1200 A/cm^2 (I_c = 9 A) was achieved for 20% silver added tapes, which is the highest in iron-based wires and tapes so far. The J_c is almost field independent between 1 T and 10 T, exhibiting a strong vortex pinning. Such a high transport critical current density is attributed to the weak reaction between the silver sheath and the superconducting core, as well as an improved connectivity between grains. We also identify a weak-link behavior from the apparent drop of J_c at low fields and a hysteretic phenomenon. Finally, we found that compared to Fe, Ta and Nb tubes, Ag was the best sheath material for the fabrication of high-performance 122 type pnictide wires and tapes.

1. Introduction

The recent discoveries of superconductivity in $LaFeAsO_{1-x}F_x$ (La-1111) and related compounds, with a highest T_c of $\sim$55 K, stimulate worldwide interest in the iron-based system [1–5], however, with much concern about the transport capabilities of such polycrystalline superconductors [6–11]. The intergranular J_cs of iron-based bulk superconductors have been studied by many groups, and J_cs of 10^3–10^4 A/cm^2 at 4.2 K and self field were reported for Sm- and Nd-1111 bulks, most of which are magnetic results [8–11]. Based on the potential applications, superconductors typically require stabilization using a normal metal cladding for reasons of electrical, thermal, and mechanical protection and, in general, need to be drawn into fine fibers [12]. Indeed, attempts at fabricating iron-based superconducting wires and tapes, through a powder-in-tube (PIT) method, have been made in the 1111, 122 and 11 series, however, no significant transport critical current has been reported [13–17]. It is presumably due to extrinsic blocking effects, such as non-superconducting phase (wetting phase or disordered phase) at grain boundaries, a dense of array of cracks and a reaction layer between sheath and superconducting core [8–11,13–15].

The K doped $A_{1-x}K_xFe_2As_2$ (A = Ba or Sr), exhibiting a high T_c of $\sim$38 K [3], seems more suitable for making superconducting wires or tapes because of the relatively low synthesizing temperature and no oxygen involved, compared with that for the RE-1111 series (RE: rare earth). However, poor transport properties in 122 wires are the principal limitation to technological applications [15]. In order to overcome this problem, we have recently found that the superconducting properties of polycrystalline $Sr_{0.6}K_{0.4}Fe_2As_2$, such as critical transition and irreversibility field, can be improved upon silver addition, probably due to a refined connectivity between grains [18]. In this letter, we introduce silver into Ag-sheathed $Sr_{0.6}K_{0.4}Fe_2As_2$ wires and tapes as a chemical addition. Transport critical currents were measured by a standard four-probe resistive method, and a high transport J_c of $\sim$1200 A/cm^2 (I_c = 9 A) at 4.2 K has been achieved in a 20% silver added tape. These results do support our earlier arguments that the absence of significant transport currents in the previous PIT wires or tapes was caused by extrinsic blocking effects [13–15].

2. Experimental details

$Sr_{0.6}K_{0.4}Fe_2As_2$/Ag/Fe composite wires and tapes were prepared by the in situ powder-in-tube (PIT) process. The details of fabrication process are described elsewhere [19]. Sr filings, Fe powder, As and K pieces, with a ratio Sr:K:Fe:As = 0.6:0.4:2:2.05, were ground in Ar atmosphere for more than 4 h using ball milling method, with the aim to achieve a uniform distribution. In order to investigate the effect of silver on critical currents, various amounts of metallic

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silver powder (0–20 wt.%) were added in the as-milled mixture. The final powder was filled in a silver tube (OD: 8 mm, ID: 6.4 mm). The composite were filled in an iron tube (OD: 11.6 mm, ID: 8.2 mm). The filled tube was swaged and drawn down to a wire of 1.95 mm in diameter. Some short samples were cut from the as-drawn wires for sintering. The as-drawn wires were subsequently cold rolled into, namely, thick tapes (0.8 mm in thickness) and thin tapes (0.6 mm in thickness). All the wires and tapes were heated at 850–900 °C for 35 h in Ar atmosphere.

Dc magnetization measurements were performed with a superconducting quantum interference device SQUID magnetometer. The microstructure was studied using scanning electron microscopy (SEM) after peeling away Ag/Fe sheath. The transport critical currents I_c at 4.2 K and its magnetic dependence were evaluated at the High Field Laboratory for Superconducting Materials (HFLSM) in Sendai, Japan, by a standard four-probe resistive method, with a criterion of 1 $\mu V\,cm^{-1}$. A magnetic field up to 10 T was applied parallel to the tape surface. The I_c measurement was performed for 3–5 samples to check reproducibility.

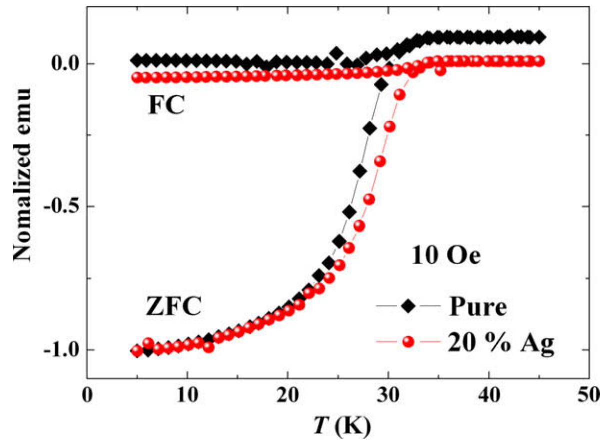

Fig. 2. Temperature dependence of DC susceptibility of the pure and 20% Ag-added tapes.

3. Results and discussion

The choice of a proper sheath was found to be critical in controlling the composition of iron-based superconducting core, and eliminating the reaction layer between superconducting core and sheath. The transverse cross-sections of a typical $Sr_{0.6}K_{0.4}Fe_2As_2$/Ag/Fe wire and tape taken after heat treatments were shown in Fig. 1a. Both Ag/Fe and $Sr_{0.6}K_{0.4}Fe_2As_2$/Ag interfaces were quite clear, indicating silver is benign in proximity to the compound at high temperatures. EDX line-scan has been performed in the direction perpendicular to the longitude of the wires and tapes. It confirms no diffusion of As or Sr into the volumes of Ag and Fe, which benefits superconducting properties of the $Sr_{0.6}K_{0.4}Fe_2As_2$ core. Most importantly, no reaction layer was observed between the silver sheath and the superconducting core (Fig. 1b). By contrast, an early attempt at fabricating $Sr_{0.6}K_{0.4}Fe_2As_2$ wires with a Nb sheath revealed a reaction layer between the sheath and the superconducting core after heat treatments, being very harmful to transport current flow [15].

The critical transition temperatures T_c of the samples were determined by the SQUID measurement. The zero-field cooled (ZFC) and field cooled (FC) magnetic susceptibility of the pure and a 20 wt.% Ag-added $Sr_{0.6}K_{0.4}Fe_2As_2$ composite tapes were measured under a magnetic field of 10 Oe (Fig. 2). It can be seen that T_c of both samples was estimated to be ~34 K, which is well consistent with that of the $Sr_{0.6}K_{0.4}Fe_2As_2$ bulks and wires prepared previously [15,18].

I_c measurements of all samples were performed using the standard d.c. four-probe method at 4.2 K in magnetic fields up to 10 T.

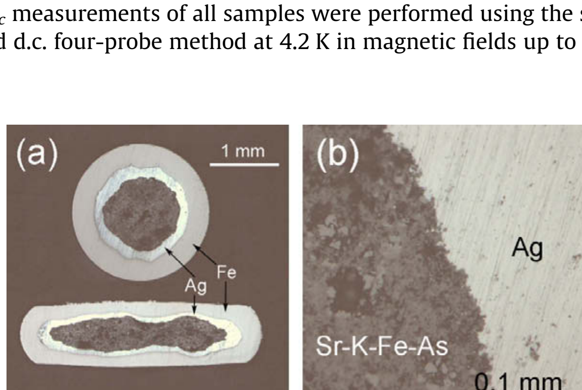

Fig. 1. (a) Transverse cross-sections of the Fe/Ag/$Sr_{0.6}K_{0.4}Fe_2As_2$ wire and a typical tape taken after heat treatment. (b) Magnified optical image of the Ag/$Sr_{0.6}K_{0.4}Fe_2As_2$ interface.

Zero resistive currents on the current–voltage curves were clearly seen for all wires and tapes. Note that the critical currents of tapes are typically higher than those of corresponding wires, probably due to higher density of superconducting core in tapes, similar to the situation for MgB_2 [20]. By contrast, we did not observe significant transport critical currents in Nb or Ta sheathed iron-based wires [13–15], which may be related to bad grain connectivity as well as the reaction layer between the sheath and the superconducting core.

Fig. 3a shows transport $J_c(H)$ curves for the thick tapes with various amounts of silver addition. Clearly, all silver added samples show a higher J_c than the pure samples in the entire field region, which is in agreement with our earlier observation that the magnetic J_c and irreversibility field H_{irr} can be improved upon silver addition in polycrystalline $Sr_{0.6}K_{0.4}Fe_2As_2$ [18]. The best J_c result was obtained by 20 wt.% silver addition, and a J_c of ~500 A/cm^2 was reached at 4.2 K and self field, a factor of ~3 higher than that of the pure samples. The almost field independent of J_c between 0.2 and 10 T suggests a strong flux pinning. A steep drop of J_c near 0.2 T was observed for all measured samples, similar to that of sintered YBCO [21], indicating a weak-link behavior.

The field dependence of J_c in an increasing as well as a decreasing field, was also characterized, and a hysteretic phenomenon has been observed. A representative normalized hysteretic J_c curve for a wire sample is shown in the inset of Fig. 3a. After increasing the field monotonically to 8 T, J_c was measured as a function of decreasing field until 0 T. Notable is the increased value of J_c in the region of 0.2–8 T, compared with the data for the virgin curve. The J_c is seen to peak on decreasing the field at 0.4 T, so that for 0 T, the J_c value is substantially reduced as compared to the virgin measurement. The hysteretic effects are supposed to be related to penetration of flux into strong pinning intragranular regions, and that the presence of intragranular critical currents enhances intergranular critical currents when the applied field is reduced from higher values [22]. This phenomenon is also a signature of weak links between grains.

Fig. 3b presents transport J_c for pure and 20 wt.% Ag-added thin tapes as a function of magnetic field. Two significant observations are recorded. First, a high critical current I_c of 9 A and 0.8 A was achieved for the 20 wt.% Ag-added tapes at self field and 10 T, respectively. The original I–V plots at 0, 1, 4, 8 and 10 T were shown in the inset of Fig. 3b. Given the average area of superconducting core (~0.75 mm^2), a very large critical current density J_c of ~1200 A/cm^2 at 0 T and ~100 A/cm^2 at high fields was obtained (Fig. 3b). These data is equivalent to the intragrain J_c of RE-1111