Research Progress in Iron-Based Superconducting Wires and Tapes

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Abstract. Nowadays, the expansion of existing electric cables has been a crucial limiting factor in the development of urban electricity power. Compared with traditional power cables, superconducting cables have advantages of large current carrying capacity, low energy consumption, and high current density. Superconducting wires and tapes, which are the main compositions of cable cores, are the basis of superconducting high-voltage applications. Among them, the 122-type iron-based superconducting wires and tapes show excellent comprehensive performance. In this paper, the development history of superconducting materials, wires and tapes and preparation method are briefly introduced, and influencing factors of critical current density of iron-based superconducting wires and tapes are summarized. Furthermore, future research direction for superconducting wires and tapes is prospected.

1. Introduction
Development of critical transition temperature of superconducting materials is demonstrated in Table 1. In 1911, Dutch physicist Onnes discovered that mercury would be superconducting near 4.15K [1]. However, the transition temperature of superconductors had only increased to 23.4K of Nb3Ge by 1986. Bodnorz and Muller [2] found in 1986 that LaBaCuO compounds showed a superconducting transition at 35K. Subsequently, Zhao Zhongxian [3] and Zhu Jingwu [4] respectively discovered Y-Ba-Cu-O oxide superconductors with Tc above 90K, indicating the critical transition temperature of superconducting materials reached liquid nitrogen temperature region (77K) for the first time.

In 2001, Japanese scientist Akimitsu et al. [5] discovered MgB2 with Tc of 39K, breaking the record Tc of alloy compound superconductor. Japan's Hosono research group [6] doped F elements in LaFeAsO compounds in 2008, which induced superconductivity and the Tc was as high as 26K. Subsequently, by applying high voltage, Tc of LaFeAsO1-xFx superconductor was increased to 43K [7]. Chen Xianhui research group [8] and Chinese scientist Zhao Zhongxian [9] discovered Sm [O1-xFx]FeAs superconductors with critical transition temperatures of 43K and 55K, respectively, which means that the critical transition temperature of iron-based superconductors broke the 40K McMillan limit temperature for the first time. In 2015, Drozdov et al. [10] studied the superconductivity of hydrogen sulfide under high pressure, and room temperature superconductivity is expected to be realized.

Table 1. Development of superconducting materials

| Tc (K) | Materials | Category |
|-------|-----------|----------|

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Superconducting Wires and Tapes

Because traditional transmission cables are restricted by small flux and large loss, it is difficult for urban electricity power to expand further. Superconducting cables have advantages of large current carrying capacity, low loss, and high current density. In addition, the total operating loss is only 50% to 60% of traditional cables while the current carrying capacity of superconducting cables with the same cross-section is 3 to 5 times larger than that of traditional cables. Hence, the application of superconducting cables can greatly save the space occupied by power transmission system, improve operating efficiency, and reduce costs.

Superconducting wires and tapes are the basis of superconducting high-voltage applications, and can be widely used in power grids, medical treatment, and transportation through high-current transmission cables and strong magnetic field coil magnets. Three important critical parameters that determine the performance of superconducting wires and tapes are: critical transition temperature (T_c), critical current density (J_c) and upper critical magnetic field (H_{c2}). The larger the three values, the wider the application range of superconducting wires and tapes [11].

2.1. Low temperature superconducting (LTS) wires and tapes

Low temperature superconducting materials are superconducting materials that have low critical transition temperature (T_c<30K) and work at liquid helium temperature. Common LTS wires mainly include NbTi and Nb_3Sn. The NbTi superconducting wire has excellent ductility and bending properties, but its upper critical magnetic field (H_{c2}) is low and current carrying capacity drops sharply under external field. Nb_3Sn superconducting wire has intrinsically higher T_c, H_{c2}, and current carrying capacity than NbTi, but its mechanical properties are poor [12], which limits its application.

2.2. High temperature superconducting (HTS) wires and tapes

High temperature superconducting materials are superconducting materials whose T_c exceeds the temperature of liquid nitrogen (77K), including four categories: 90K rare earth-type, 110K bismuth-type, 125K thallium-type, and 135K mercury-type. Thallium-type and mercury-type contain environmentally hazardous elements and the special preparation process limits their applications. Therefore, RE123, Bi2212 and Bi2223 form the three practical high temperature superconducting material system [12].
2.2.1. The first generation of high temperature superconducting wires and tapes. 1G-HTS tapes based on Bi2223 and Bi2212 are obtained through traditional powder-in-tube and wire drawing process. Bi2223 tape has mature preparation technology, large current carrying capacity and high Tc, but its irreversible magnetic field at 77K is very low (only 0.2T), and its critical current density (Jc) declines rapidly with the magnetic field. Moreover, silver is used for coating, which is costly. Bi2212 can be made into a round line shape so problems caused by anisotropy can be ignored. However, shortcomings such as low critical current density, high void rate of finished product, poor mechanical properties, and difficulty in price reduction, limit its application [12].

2.2.2. The second generation of high temperature superconducting wires and tapes. 2G-HTS tape based on RE123 was developed through film epitaxy growth technology and biaxial texturing technology. The price of 2G-HTS tapes raw materials is lower than that of 1G-HTS, and cheaper Ni-based alloy can be used [12]. Although the critical transition temperature has exceeded the liquid nitrogen temperature of 77K, the superconducting coherence length (ξ) of 2G-HTS is very small. Large crystal defects will become weak connections and some effective magnetic flux pinning centers in conventional superconductors are no longer effective, resulting in weak flux pinning ability and obvious magnetic flux creeping. Thus, weak magnetic flux pinning ability and weak connection of grain boundaries both greatly limit its critical current density. When the grain boundary angle of YBCO increases from 3° to 45°, the weak connection effect is obviously shown, and the current carrying capacity of grain boundary drops sharply by about four orders of magnitude, from 10⁶ to 10⁷ A/cm² to 10² to 10³ A/cm². In order to reduce the impact, biaxial texturing process must be applied [13]. However, the technical steps of this process are complicated, leading to very high production costs.

2.3. Iron-based superconducting wires and tapes
Iron-based superconducting materials can be roughly divided into the following four types shown in Table 2 according to the composition ratio and crystal structure of parent compound [14]:

| Types    | Crystal structure [15] |
|----------|------------------------|
| 11-type  | FeSe (Te)              |
| 111-type | AFeAs (A=Li, Na)       |
1111-type LnOFePn (Ln=La, Ce, Pr, Nd, Sm, Gd, Tb, Dy, Ho, Y; Pn=P, As) and DvFeAsF (Dv=Ca, Sr) etc.

122-type AFe$_2$As$_2$ (A=Ba, Sr, K, Cs, Ca, Eu) etc.

1111-type has the highest Tc in the entire iron-based superconducting system, but its doping element is basically F element, which is easily lost during high temperature heat treatment, causing the increase in complexity of synthesis and control of its uniformity [16]. Moreover, the anisotropy of 1111-type is the largest ($\gamma_H=2$–$5$), the higher the value of anisotropy, the lower the working temperature of superconductor. Although 11-type has the simplest structure, its Tc is very low (8K) and its application range is limited to liquid helium temperature zone. 111-type iron-based superconductor is unstable in the air, so there are few researches on it [11]. 122-type has Tc of about 38K, which can be operated in the medium temperature region without relying on liquid helium [11]. In addition, its anisotropy is weak, upper critical field and critical current density is high [17], attenuation under magnetic field [18], requirement for coating materials, and price is relatively low. Therefore, 122-type iron-based superconducting material exhibits excellent comprehensive performance among the existing superconducting materials in the application field of low temperature and high field.

In terms of high-field applications, the upper critical magnetic field of 122-type iron-based superconductor is still as high as 70T even at a temperature of 20K [11]. At low temperatures, the 122-type and 11-type iron-based superconductors are isotropic [19]. Although anisotropy of superconductors increases with higher temperature, the anisotropy of iron-based superconductors is about 3 when the temperature is close to Tc, which is still smaller than the anisotropy of ReBCO coated conductors ($\approx$5) and Bi-based superconducting tapes ($\approx$50). Consequently, the magnetic flux pinning ability of iron-based superconductors is stronger than that of copper-based oxide superconductors [20], and its pinning potential decays slowly as the applied magnetic field increases.

For critical current density, iron-based superconductors have a very strong intrinsic critical current [21], and are insensitive to doping, that is, can tolerate more defects. Hence, the magnetic flux pinning ability of iron-based superconductors can be improved by introducing appropriate defects, which act as effective pinning centers. The critical current density of iron-based superconductors under magnetic field can be effectively increased by the introduction of a large number of pinning centers [22]. The coherence length of iron-based superconductors is relatively short, about 1.8–2.4nm [23]. Furthermore, unlike traditional low temperature superconducting materials (such as NbTi or Nb$_3$Sn materials), iron-based superconducting materials have anisotropic layered structure, so they may have an intrinsic factor, the weak grain boundary connection effect, that limits the critical current density [24]. The current carrying performance is mainly restricted by grain boundary in iron-based superconducting wires and tapes with weak connectivity [23]. When the grain boundary angle ranges from 9° to 45°, the critical current density of iron-based superconductor decreases by an order of magnitude, and almost remains
stable within 9°. Thus, its critical angle of weak grain boundary connection effect is about 9°, which is larger than 3~5° in YBCO superconductor [19]. Therefore, weak grain boundary connection effect of iron-based superconductor is smaller than that of copper oxide superconductor, leading to relatively smaller attenuation of superconducting current flow through the large-angle grain boundary and stronger tolerance to grain boundary angle.

For coating material and cost aspect, compared with bismuth-based oxide superconductor which is also prepared by powder-in tube process, 122-type iron-based superconductor contains no oxygen, so is not restricted by the oxygen permeability of coating material. Hence, 122-type iron-based superconducting wires and tapes have a wider choice of coating materials, except for silver, cheaper materials such as copper, iron, stainless steel can be selected, which can greatly reduce the manufacturing cost [11].

3. Preparation Methods of Iron-based Superconducting Wires and Tapes
Iron-based superconducting wires and tapes have a great prospect in the application field of low temperature and high field. Due to the difficulty in plastic processing, powder-in tube method (PIT method) is usually used for preparing iron-based superconducting wires and tapes [25]. The main preparation steps of PIT method are illustrated in Figure 1: (a) The raw materials are repeatedly calcined and ground, then the obtained powders are filled into metal tubes (the main component is Ag). (b) Metal tubes are subjected to deformation processing such as drawing and swaging, then assembled again into metal tubes and drawn, and finally repeatedly rolled and calcined to form superconducting tapes. (c) In order to eliminate the micro voids formed in raw materials and improve the current carrying capacity, formed tapes need to be processed under high pressure to finally form the finished tapes [26, 27].

According to the difference between precursors, PIT method can be divided into In-situ method that uses uniformly mixed initial powders, firstly cold-formed and then sintered into phase, and Ex-situ method that uses superconducting powders, firstly sintered into phase and then cold-formed. The preparation procedure of In-situ method is simple, but the sintered superconducting core usually contains a large number of impurity phases, and requirements for the chemical stability of coating materials are extremely high since avoiding chemical reactions with raw materials during heat treatment is necessary [16]. Besides, problem of low density is existed [23]. According to the reported results, when the iron-based superconducting wires and tapes were prepared by In-situ method, voids that seriously affect the critical current density would be produced [16].

Ex-situ method usually has a one-step annealing process after forming, which can effectively improve grain connectivity and avoid voids that generated during the chemical reaction in In-situ method [16]. In order to improve the phase purity of precursor powders, grinding and sintering steps can be repeated many times. At present, Ex-situ method has become the main method for preparing iron-based superconducting wires and tapes with high performance [25].
Figure 1. Procedure of preparing iron-based superconducting wires and tapes by PIT method [11]

4. Influencing Factors for Critical Current Density of Iron-based Superconducting Wires and Tapes

Factors such as voids, impurity phases, and amorphous layers in the material will inhibit the order parameters of grain boundaries and thus limit critical current density. For iron-based superconducting wires and tapes, critical current density could be effectively increased by improvement of coating materials, chemical doping and quality improvement of precursor powders [28].

4.1. Coating materials of iron-based superconducting wires and tapes

For PIT method, the metal coating material should have good mechanical properties, a melting point higher than the sintering temperature, and not react with the superconducting core at high temperature. In order to enhance the phase purity of iron-based superconducting wires and tapes, a high sintering temperature is usually required. As shown in Table 3, Nb, Ta, Fe, and Ag all have high melting points [29,30], but Nb and Ta are soft and difficult to be drawn into wires. Besides, after high temperature sintering, Nb and Ta will react with the superconducting core to form Nb, Ta arsenide reaction layer, which completely hinders the flow of current from coating material into superconducting core [31]. Fe has good mechanical properties and the superconducting core drawn by Ex-situ method is relatively uniform, which is also easy to form rolling texture. However, Fe will also react with the superconducting core at high temperature to form an iron arsenide reaction layer [32].

Ag hardly reacts with the superconducting core below the melting point, which is conducive to the crystal growth of iron-based superconductors and formation of reaction layer is avoided [33]. In general, Ag is an ideal coating material for iron-based superconducting wires and tapes.

Table 3. Property comparison of coating materials for iron-based superconducting wires and tapes

|                | Nb                                | Ta                                | Fe                                | Ag          |
|----------------|-----------------------------------|-----------------------------------|-----------------------------------|-------------|
| Mechanical property | Difficult to be drawn into wires | Difficult to be drawn into wires | Uniform wires, easy to form rolling texture | Good ductility |
| Melting points (°C) | 2468                             | 303.5                             | 1538                              | 961.78      |
| Reaction with core | Nb arsenide reaction layer        | Ta arsenide reaction layer        | Fe arsenide reaction layer        | Hardly react |

4.2. Quality improvement of precursor powders for iron-based superconducting wires and tapes

Togano et al. [34] used a high-temperature melting method to prepare precursor powders. Different from the traditional ball-milling process, the heat treatment of the high-temperature melting method was performed at a high temperature higher than the melting point of FeAs compound (1050°C) (heated in a box furnace at 1100°C for 5 minutes) to fully react and mix the components. The prepared precursor powders had good grain connectivity and no FeAs phase. Figure 2 demonstrates that Jc of Ag coated Ba0.6K0.4Fe2As2+Ag wire prepared by the high-temperature melting method reached 10^6 A/cm^2 and 1.1 × 10^5 A/cm^2 at 4.2K zero field and 10T, respectively.
4.3. Chemical doping of iron-based superconducting wires and tapes

The superconducting core of iron-based superconducting wires and tapes is polycrystalline material, and chemical doping is one of the most effective approaches to promote the superconducting properties of polycrystalline materials. Chemical doping can introduce magnetic flux pinning center, promote the generation of superconducting phase, and improve the coupling between grains [15].

Wang et al. [35, 36] studied the influence of Ag doping (0-20 wt%) on polycrystalline Sr$_{0.6}$K$_{0.4}$Fe$_2$As$_2$ superconductors. The experimental results indicated that Ag doping did not enter the grain boundary, and effectively inhibited the formation of amorphous phases and layers. In Figure 3 and Figure 4, larger grains could be observed and some had clear boundaries, with most of the grain boundaries were tightly connected. After Ag doping, the critical transition temperature of the sample did not decrease while the critical magnetization current density was greatly improved, and the sample possessed high density and less amorphous phase. The improvement of superconductivity of the sample might be caused by better connectivity between larger crystal grains. Figure 5 shows that the critical current density of the Sr$_{0.6}$K$_{0.4}$Fe$_2$As$_2$+20% Ag tape at 5K temperature and 6.5T magnetic field reached $1.5 \times 10^3$ A/cm$^2$. 

![Figure 2. Curves of Jc and magnetic field of wires heat-treated at 850°C for 3, 15, 30h [34]](image)

![Figure 3. TEM image of un-doped sample [36]](image)
Figure 4. TEM image of sample doped with Ag [36]

Figure 5. The critical current density (\(J_c\)) caused by the hysteresis of sample at 5K and 20K [36]

During the deformation processing of iron-based superconducting wires and tapes, a mass of superconducting crystal grains would fracture, resulting in poor connectivity and weak transmission performance. Sn element has a low melting point (232°C) and can act as a good metal flux for growth of 122 phase. During high temperature heat treatment, Sn can promote crystallization at grain boundaries, reduce the formation of amorphous layers, and thereby improve inter-crystalline connectivity.

Gao et al. [37] prepared Sn+Sr0.6K0.4Fe2As2 tapes. Figure 6 illustrates that the transmission \(J_c\) under 4.2K zero field and 10T were as high as \(2.5 \times 10^4\) A/cm\(^2\) and \(3.5 \times 10^3\) A/cm\(^2\), respectively. Though \(T_c\) of Sn-doped samples dropped by about 1~2K, the transmission performance of Sr122+Sn tape was further promoted by improved heat treatment process [38]. The Sn-doped sample had fewer voids and micro-cracks, and good crystal grain connectivity with crystal grains tended to be pancake-like. The excellent \(J_c\) performance might be due to the good texture of grains and contact morphology of grains in the superconducting core changed by Sn, which helped form new superconducting phases at grain boundaries, thereby the interface energy was reduced and inter-grain coupling was enhanced. Moreover, similar to Ag doping, no amorphous layer was formed at grain boundaries of the Sn-doped sample. It should be noted that Sn element could shorten the reaction time of superconducting materials so that
wires and tapes with good connectivity performance could be formed within 1h while the heat treatment time in un-doped samples and Ag-doped samples were generally 20~30h [38].

![Figure 6. Jc of un-doped and Sn doped samples under different magnetic fields at 4.2K [37]](image)

5. Future and Prospect

The practical application of superconducting cables still has a lot of room for improvement. The first one is to improve the existing preparation method of superconducting wires and tapes and promote the performance of low-temperature refrigeration devices, thereby reducing the cost of superconducting cables. The second one is to explore new superconducting materials that are more suitable for practical applications. Alkali metal, rare earth, transition metal and C, N, O, F elements are predicted to be the elements required for new high-temperature superconductors.

Although the transition temperature of iron-based superconducting material, which is 38~55K, has not reached the temperature region of liquid nitrogen, its upper critical magnetic field is as high as 70T at 20K, and 122-type iron-based superconducting materials also have advantages of high critical current density, weak anisotropy, low requirements for coating materials, slower attenuation under the magnetic field, and low-cost PIT preparation method, making iron-based superconducting material a new potential superconducting material in the application field of 4.2~30K low temperature and high field in the future.

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