Towards an ideal world with superconductivity

—Current status and prospects for rare-earth barium copper oxide superconducting tapes—

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We review the history, current status, and prospects of research on RE $Ba_2Cu_3O_y$ (RE: rare earth element) coated conductors. Three major issues were addressed to achieve critical current performance for long-coated conductors of several hundred meters. Special functional performances, e.g., in-field critical current, were greatly improved. Applications of coated conductors were also initiated. We expect applications to appear in the near future.

Keywords : Superconductivity, rare-earth barium copper oxide (RE $Ba_2Cu_3O_y$) coated conductors, critical current density, artificial pinning centers, AC loss

1 Introduction

Superconductivity, discovered in mercury by Heike Kamerlingh Onnes of the Netherlands in 1911, was found in the process of creating liquefied helium.[1] Superconductors have been expected to be dream materials, since superconductivity allows transfer of electric energy at zero resistance.

In superconductivity, there are three critical conditions: critical temperature (T_c: the maximum temperature at which superconductivity is revealed); critical current density (J_c: the limit value of current per unit cross-sectional area at which superconductivity can be maintained); and critical magnetic field (H_c: the limit value of magnetic fields at which superconductivity can be maintained). Among these three conditions, T_c and H_c are physical properties that are essentially determined by the material. There are two types of superconductors, type I and type II. In a type I superconductor, superconductivity immediately fails when H_c is reached at relatively low magnetic field, while a type II superconductor has two H_c values, and is able to maintain superconductivity to relatively high critical field (H_{c2}) as some of the magnetic flux infiltrates inside the superconductor when the magnetic field surpasses the lower critical field (H_{c1}). Therefore, in practice, the materials with high T_c and H_{c2} are selected for the type II superconductor, and development has been done to achieve high J_c. As a result, NbTi and Nb_3Sn are now being used for magnetic resonance imaging (MRI), MAGLEV, and others. However, since T_c is 20 K or less, these materials must be kept in liquid helium (boiling point 4.2 K). Liquid helium is obtained as a by-product of natural gas, and in Japan, it is mostly imported. However, as the energy source shifts from natural gas to shale gas, the price has skyrocketed and has become very expensive, and the supply is becoming unstable. Moreover, since it is ultra-low in temperature, extremely low specific heat is another problem. The heat generated or entered by accidental thermal agitation easily raises the temperature, and the temperature may surpass T_c and cause a phenomenon called quench in which superconductivity is rapidly lost. Both are issues that arise from ultra-low temperature, and the discovery of materials that show superconductivity at high temperature was long awaited.

As the search continued, J. G. Bednorz and K. A. Müller of Germany discovered a superconductor of a new material with high T_c (high-temperature superconductor) in 1986.[2] While the previous superconductors were all metal materials, the material they discovered was an oxide $La_{2-x}Ba_xCuO_4$. When they found this material, they were not looking for superconductors but were actually in the process of developing a conductor. In this material, the temperature at which zero resistance was reached was about 10 K, and it was not so high compared to the metal materials. Therefore, it did not make news at the time of its discovery. However, Professor Shoji Tanaka of Tokyo University (at the time) focused on the point that the temperature at which the resistance began to fall was over 30 K, and by surveying the materials that had similar composition to this material, he discovered a new superconductor that surpassed the T_c limit as predicted by the Bardeen-Cooper-Schrieffer (BCS) theory.[3] Since then, new superconductors were found, and some of them had T_c that surpassed the boiling point of liquid nitrogen (77 K),[4]–[6] and there was expectation for wider-ranging applications past the

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Table 1. Main elemental technologies corresponding to issues

Issue	Elemental technology	Breakthrough technology
Development of fundamental technology for high J_c (1st issue)	・Fabrication technology for textured substrate/buffer layer Ion beam assisted deposition (IBAD) Textured metal substrate ・Reaction control technology for superconducting layer/metal substrate ・Control technology for impurities in superconducting layer (single-phase technology) ・Technology for flattening metal substrate (defect control technology) ・Technology for electric and chemical stabilization	・Large surface area film-forming ・Self-texturing technology ・High-speed texturing material
Development of high-performance long-length tape (2nd issue)	・Technology for fabricating long-length superconducting layer with high *Ic* property Pulsed laser deposition (PLD) Metal organic deposition (MOD) Metal organic chemical vapor deposition (MOCVD) ・Technology for uniform properties (control of composition, film thickness, defects, etc.) ・Development of technology for high mechanical strength ・Development of low-cost technology	・Multi-plume and multi-turn system ・Substrate temperature control ・Reaction mechanism analysis ・Preparation composition control ・Gas flow control
Development of technology to improve specific performance for application (3rd issue)	・Control technology for artificial pinning center (technology to improve in-field J_c) ・High-precision scribing technology (narrow tape processing, damage-less, high speed, etc.) ・Achievement of high engineering critical current density (achievement of thin metal substrate) ・Tape with isotropic properties (low aspect ratio, wire, achievement of isotropic J_c (B)) ・Superconducting joint technology with low resistance	・Fine artificial pinning material ・UTOC(Ultra-thin Once Coating)-MOD method ・Scribing technology with excimer laser

conventional limitations. Among these new superconductors, RE $Ba_2Cu_3O_y$ (REBCO; RE or rare-earth elements including yttrium) with T_c up to 95 K and $Bi_2Sr_2Ca_2Cu_3O_x$ (BSCCO) with T_c up to 110 K were targeted for development as industrial materials, as they had high T_c and did not contain toxic elements.

To use these superconductors as industrial materials, they must be processed into tape or wire forms that allow them to be made into devices. Here, the issue was that the subject superconductor was an oxide. In conventional metal materials, it is possible to manufacture long and even wire by using drawing techniques, but similar methods cannot be used with oxides because of poor ductility. Therefore, wire-making was developed for BSCCO prior to REBCO. Since BSCCO materials have a relatively good sliding property between the crystal grains, wire can be formed by using the modified drawing method. Long-length wire with advanced properties has been successfully manufactured by a method called a silver sheath method, where a silver pipe is filled with raw material powder, and this is repeatedly rolled out, heat treated, and grouped.[7][8] On the other hand, this material had issues in mechanical properties as well as in in-field J_c properties at relatively high temperature (up to 77 K). To solve these issues, the development was started in earnest for a REBCO material with excellent J_c properties in high temperature and magnetic fields. National projects and development were started almost at the same time in Japan and the USA around 2000.

The development of a REBCO material was conducted by solving three main issues. The first issue was the development of technology to form a biaxially oriented structure (structure in which all crystal axes are oriented uniformly as in single crystals) that was necessary to achieve high J_c properties of REBCO in a tape form. The second issue was the development of manufacturing long-length tapes using the technology of forming long-length thin films with high I_c properties (critical current that is the limit value of currents that allows superconductivity to be maintained; J_c × cross-section surface area). To achieve high I_c, the technology for forming a thick film while maintaining high J_c properties was essential. Here, the product of $I_c \times L$ was used as an index, and technological development was conducted on how to achieve high I_c properties in a long-length tape. The product of $I_c \times L$ not only is an index that showed both the superconducting properties (I_c) and length (L) that are necessary elements for a long-length material, but is equivalent to the magnetomotive force (ampere-turn) when a coil is formed. After solving the second issue, development of devices using long-length tapes was started, and the requirements for tapes shifted to specific properties and functions that corresponded to use in specific devices. As the third issue, we are currently working on the technological development to increase specific performances such as in-field I_c properties and low-loss tapes. Figure 1 shows the concept from inception to realization including the development issues, and Table 1 summarizes the elemental technologies for each issue.

In this paper, we outline the R&D conducted to solve each of the aforementioned issues through the selection of key technologies listed as the elemental technologies in Table 1. Here, the results of development at the International Superconductivity Technology Center (ISTEC) at which the author conducted research and AIST with which the author is currently affiliated will be featured. ISTEC was established in 1988 after the discovery of the aforementioned high-temperature superconductor. It is an incorporated foundation whose purpose was the promotion of R&D for high-temperature superconductors and their dissemination through activities such as organizing academic conferences. ISTEC was a joint industry-academia-government research center consisting of affiliated researchers, researchers dispatched from private companies, and foreign researchers from abroad. It operated for about 30 years until it was dissolved in 2016, and during its operation, it received subcontracts for several national projects from the Ministry of Economy, Trade and Industry (METI) and the New Energy and Industrial Technology Development Organization (NEDO). It formed joint research units with several private companies, universities, and national research institutes, to lead the world in the development of high-temperature superconductor technology. The strategy shown in Fig. 1 was created mainly by ISTEC, and was later shared in the world.

Representative national projects include, in chronological order: "R&D Project for Core Technology of Superconductivity Application" Phase I and Phase II, "Technological Development of Yttrium Superconducting Electric Devices," "Technological Development of Yttrium Superconducting Electric Device (Joint Core Technology Development)," and others. These projects were subcontracted by METI, NEDO and the Japan Agency for Medical Research and Development (AMED). The early efforts centered on the development of tapes, and the latter shifted toward development of devices. ISTEC took lead in forming the joint research units with companies, universities, and national research institutes, and then executed the developmental projects. It has become part of AIST and participates in the "Technological Development to Promote Practical Use of High-Temperature Superconductivity," and continues the technological development to advance tape materials.

2 First issue (Development of high J_c core technology)

REBCO has the potential of having excellent J_c properties, but it is also known to be greatly dependent on the orientation of the crystal grains (the direction in which the crystals are arranged). Therefore, the first major issue was how to achieve biaxial orientation in tape formation, and various approaches were taken to realize this. First, wire-making was done using the silver sheath method that was successful for BSCCO material. Although it was possible to achieve a wire form using this method, it was difficult to achieve biaxially orientated crystal grains, and due to cracks and other issues, the best we could do was a low J_c property of 10^3 A/cm^2 achieved in a self-generated magnetic field (a magnetic field generated by running current through linearly arranged wires) in liquid nitrogen (77 K).[9] In practice, it is thought that a J_c property of at least 10^5 A/cm^2 (77 K, self-generated magnetic field) is necessary, and dramatic improvement was necessary. Later, several methods that enabled biaxial orientation was developed and this led to dramatic improvement of the properties. In a layered structure (Fig. 2) in which a number of intermediate layers were added to the metal substrate, these methods realized biaxial orientation by intermediate layers made by a special film-forming method or special treatment of the metal substrate. In this paper, the ion beam assisted deposition (IBAD) method, which has been used most widely around the world and is becoming

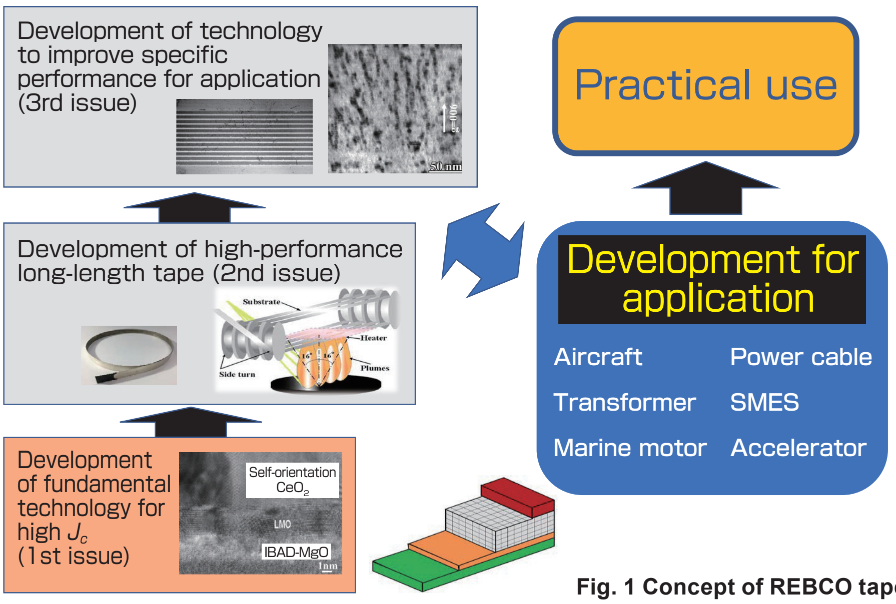

Fig. 1 Concept of REBCO tape material

the mainstream in Japan, will be explained.[10][11] Basically, this is a method in which a film is formed by hammering film-forming deposition seeds toward a metal substrate by irradiating ion beams to the bulk body of a target substance, and then depositing them on the substrate. The IBAD is a method in which another ion beam is shot onto the metal substrate during film-forming at a certain angle (Fig. 3). The angle differs according to the material, and it is 55º from the normal direction for ZrO_2 that was a material used initially. It is 45º for MgO that is recently used, and in-plane orientation can be obtained. These angles follow the close-packed surfaces of materials against the biaxially oriented structure of the substrate, and an ion channeling mechanism (mechanism by which the orientation structure is formed by vapor deposition by limiting the lattice that allows the direction of transmittance using the frame formed by ion beams aimed at the substrate) and a bombarding mechanism (mechanism for forming the orientation structure by kicking out crystals that are not oriented in certain ways using ion beams aimed at the substrate) are proposed.[11] This method is effective in achieving a high degree of biaxial orientation on a substrate without orientation, and J_c properties surpassing 1 MA/cm^2 were achieved at 77 K self-generated magnetic field for the first time for a REBCO superconducting tape on a metal substrate. On the other hand, since the method involves the selection of film-forming deposition seeds, the manufacture speed of the tape is slow (initially < 1 m/h) because the film-forming speed is slow, and this was a major issue. To solve this issue, ISTEC attempted the following two methods.

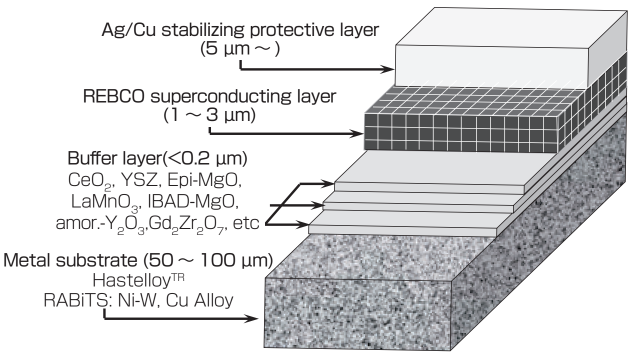

Fig. 2 Schematic diagram of REBCO tape structure

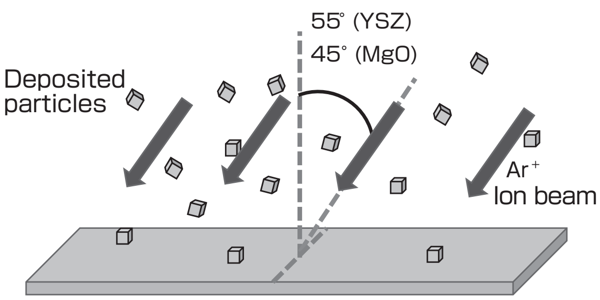

Fig. 3 Conceptual diagram of IBAD method

One was the method of increasing the deposition surface area and increasing the moving speed by increasing the number of film-forming turns. This was conducted through the development of a device in a national project. Second was the discovery of a new phenomenon to improve the degree of orientation. This was the phenomenon called "self-orientation" in which the orientation speed dramatically increases when CeO_2 is film-formed by a usual physical vapor deposition method without an assist beam with a high rate, after giving a certain degree of biaxial orientation by the IBAD method.[12] Self-orientation can be seen in other materials, but the presence of an IBAD layer as a base is a requirement, and the orientation mechanism is being clarified.[13]

In addition, it was found that MgO was suitable for speeding up the IBAD method in a study in the USA.[14] ISTEC incorporated this result quickly in Japan. While it required about 3 hours to get the in-plane orientation (Δφ) to 10º or less by a conventional IBAD method, the in-plane orientation of Δφ–5º could be obtained in a few minutes by combining the aforementioned two methods (Fig. 4). The manufacture speed of 50 m/h was surpassed, and it is no longer a speed-limiting process. Also, the improved orientation increased the J_c value for which several MA/cm^2 (77 K, self-generated magnetic field) was surpassed. Critical current (I_c) of several hundred A/cm was achieved with film thickness of 1 µm, and the stage of demonstrating the attractiveness as a tape material was reached.

3 Second issue (Development of high-performance long-length wire)

As described above, the biaxially orientated structure necessary to attain high J_c for the REBCO tape material was achieved by solving the first issue. In this chapter, we explain the development of film-forming technology for superconducting layers that are formed on this oriented substrate.

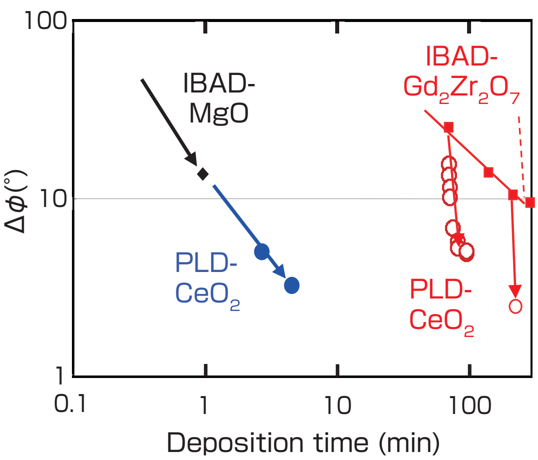

Fig. 4 Self-texturing phenomenon on IBAD layer

The technologies to form the superconducting layers are roughly divided into gas phase methods and liquid phase methods. The gas phase methods include pulsed laser deposition (PLD) and metal organic chemical vapor deposition (MOCVD), and a representative chemical liquid phase method is the metal organic deposition (MOD) method. At ISTEC (AIST), development was conducted by PLD and MOD methods using trifluoroacetate (TFA) as the raw material. The following is an account of the characteristics of the two methods and their major results.

The PLD method is a method by which excited deposition seeds are deposited onto a substrate material by irradiating the target material with an excimer laser. Since the film is formed in vacuum, impurities are unlikely to become included, and a high-quality film can be formed easily. Since the energy density of the excimer laser is high, the transferability from the target to the film is high. Therefore, it is effective on materials with which complex composition must be controlled carefully, and this method is suitable for RE superconducting materials that are the target of this study. In fact, we have been successful in forming a relatively high-quality film from the early stages of tape development, and the J_c properties were improved along with the improvement of the biaxial orientation of the intermediate layer. On the other hand, since the device is expensive, the cost of tape may increase. To solve this issue, it was thought necessary to improve the yield and manufacturing speed, as well as improving the properties. Against such a background, a multi-plume and multi-turn method was developed in a reel film-forming method at ISTEC. Plume is a bunch of deposition seeds excited by a laser, and they rise up from the target like flames. To realize high-speed film-forming, a large laser (for example 200 W) is utilized to supply large amounts of excited deposition seeds to the substrate with large power. When this is done, the degree of supersaturation becomes too large with one plume, epitaxial growth in the oriented intermediate layer cannot be maintained, and a superconducting film with biaxial orientation cannot be achieved. Therefore, we developed a multi-plume method in which pulsed laser irradiation locations are dispersed, several plumes are created, and supply volume from the plumes can be controlled (Fig. 5).[15] This allows the control of the degree of supersaturation, and high speed was achieved while maintaining high J_c in the oriented film. On the other hand, the multi-turn method is a way for achieving high speed and high yield. The aforementioned plume spreads from the target and travels toward the substrate. The spread of the plume is larger than the size of the substrate (up to 10 mm), and the raw materials are lost in areas outside the substrate when film-forming is done on a moving substrate. Therefore, we introduced a multi-turn system where the substrate is turned by sliding the tape. By doing so, the raw material can be recovered from a wide area, and we were able to obtain the movement speed necessary to form the same film thickness.

There was also the issue of achieving high I_c. As described above, the PLD method is a method suitable for high J_c. It is certainly possible to obtain high J_c in an area in which film thickness is thin, but in general, the J_c decreases as the film thickness increases, and this was a barrier to achieving high I_c. For this issue, we hypothesized that the radiation rate increased due to decrease in surface flatness as the film thickness increased, and as a result, the surface temperature decreased. In our PLD device, heating is done from the backside of the tape, and the temperature of the film is determined by the balance of incoming heat and heat radiation from the superconducting film surface. Therefore, we thought that the surface temperature decreased as the radiation amount increased by the increased heat discharge of the film surface. As countermeasure, by film-forming in a temperature environment that is controlled in a pattern in which substrate control temperature is increased according to the increase in film thickness, we succeeded in maintaining high J_c during formation of a thick film.[16] As a result of these high speed and high property achievements, we succeeded in the manufacture of tape material with 500 m length and 300 A/cm width, at a relatively early stage (Fig. 6).[17][18]

The MOD method is a process of forming a superconducting film by coating with a raw material solution and heat-treating in an electric furnace. It is known as a low-cost process since

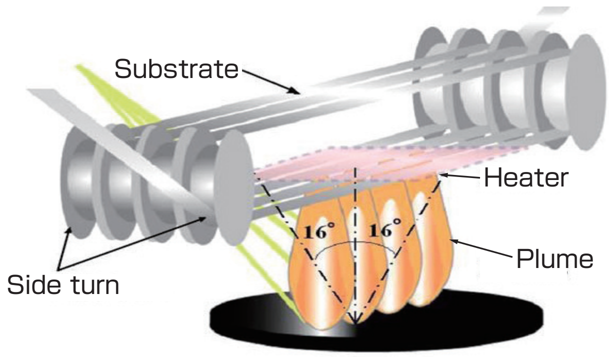

Fig. 5 Conceptual diagram of multi-plume and multi-turn PLD system

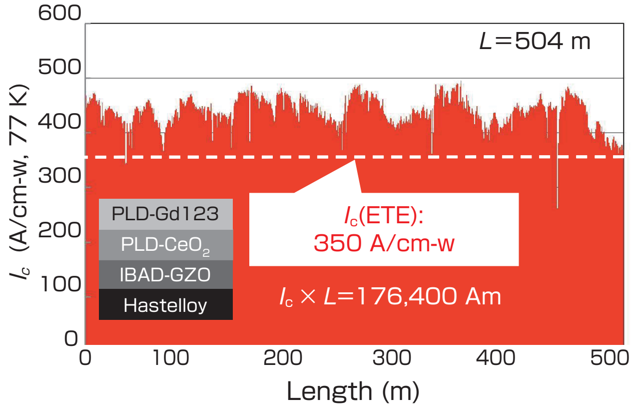

Fig. 6 Long-length tape in IBAD-PLD tape material[18]

it does not require an expensive vacuum chamber or a heat source. However, in a general MOD method, the reaction is determined by pyrolysis, and epitaxial growth is difficult to obtain if there is no temperature difference in the film, and it was unsuitable for a REBCO superconducting film that was our target. However, large progress was made as epitaxial growth was made possible by using BaF_2 as an intermediate product while using TFA as a raw material.[19] For the formation of superconducting layer in this system, as shown in Equation (1), it is necessary to supply water to BaF_2 and have them react.

$$\frac{1}{2}Y_2Cu_2O_5(s) + 2BaF_2(s) + 2CuO(s) + 2H_2O(g)$$
$$\rightarrow YBa_2Cu_3O_{6.5+x}(s) + 4HF(g) + \frac{x}{2}O_2(g)$$
… Equation (1)

In this reaction, HF gas is generated as a reaction product (Fig. 7). This conversion reaction enables epitaxial growth. We first analyzed the reaction mechanism, and found that the growth rate is determined by the exhaust speed of HF gas (V_g) and steam partial pressure (P_{H2O}) and total pressure (P_t) and others.[20]

$$R \propto \frac{\sqrt{V_g}\sqrt{P_{H2O}}}{P_t}$$
… Equation (2)

In this method, improvement of I_c was an important issue, and technological development was aggressively pursued. A representative method is to achieve high properties by controlling the starting composition. In the MOD method, it was initially difficult to achieve complete reaction during epitaxial growth, and the intermediate products ($Y_2Cu_2O_5$, BaF_2, CuO, etc.) were often incorporated into the superconducting layers. In that case, while $Y_2Cu_2O_5$ and CuO became spherical and seldom became current inhibiting factors, Ba compounds tended to be present in the grain boundary, easily deteriorated, and were thought to cause the deterioration of properties. Therefore, we succeeded in achieving stable high properties by changing the starting composition to Ba deficient.[21] Also, in the MOD method, the property surface does not change greatly since the reaction progresses from the bottom of the calcinated film that is used as the precursor, there is no temperature change by thickness as seen in the PDL film, and the decrease of J_c is not likely to occur against the film thickness. Therefore, we were able to achieve high I_c by thickening the film.[22]

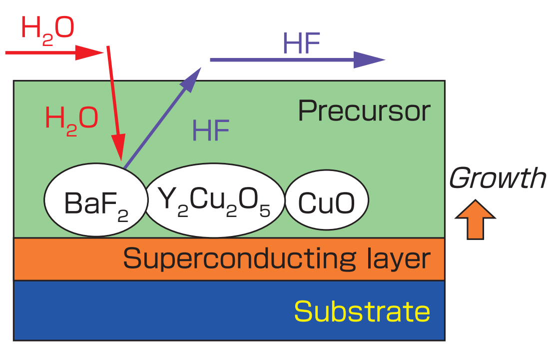

Fig. 7 Conceptual diagram showing reaction in TFA-MOD method

For achieving long length, development was conducted by two major processes. One was a method called a reel-to-reel (RTR) method in which the tape with precursor film supplied from a reel is fed into an electric furnace to undergo heat treatment. This method allows stable reaction control as soon as a certain steady state is established, and therefore is a method suitable for stabilizing the long-length properties. However, there is an issue of manufacturing speed. The other is the batch method. This is a method where the precursor film is rolled onto a drum and heat-treated inside a large furnace. While it has excellent manufacturing speed, the issue is how to reduce the location dependency of the growth environment (temperature, gas flow, etc.). Both methods were optimized by controlling the temperature pattern and gas flow based on the basic investigations described above, and we succeeded in manufacturing a high-property long-length tape as shown in Fig. 8.[18][23]

The long-length tape was achieved through joint development by ISTEC and tape manufacturers. The PLD method was a joint research mainly with Fujikura Ltd., and the TFA-MOD method was developed with SWCC Showa Cable Systems Co., Ltd. The results were obtained in the NEDO projects. On the other hand, a large-scale national project was done to develop tapes in the USA, and as a result, Japan and USA became leaders in the development of RE superconducting tapes. Here, development was conducted by setting as index the product of $I_c \times L$ that shows both the properties and length at the same time. Figure 9 shows the change in the $I_c \times L$ product.[24] Rapid progress was seen at around 2000 when

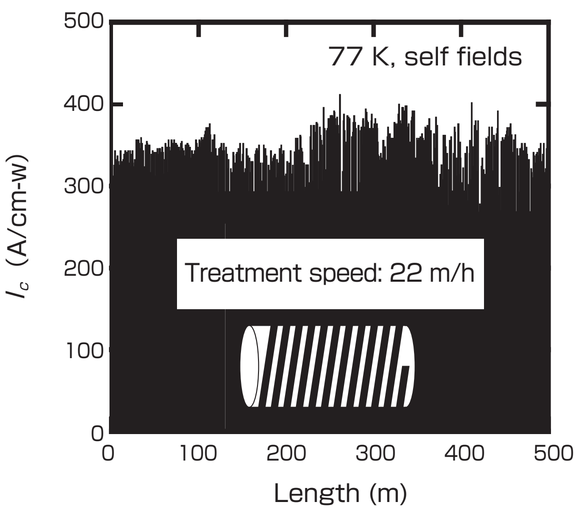

Fig. 8 Long-length tape in IBAD-MOD tape material[18]

the national projects were started in Japan and the USA. It can also be seen that it was led by researchers of Japan and the USA. The Japan-USA competition affected not only the academic competition through international conferences, but also budget procurement that was affected by the difference in the start of the fiscal year between the two countries. As a result, this generated great synergy, and dramatic progress was seen. Later, SuNAM Co., Ltd. of Korea and partially Russia-financed SuperOX joined the race. They are now selling tape products with several hundred A of I_c at several hundred m length.

4 Third issue (Technology to improve specific performance for use in devices)

As it became possible to manufacture tape material with a certain performance at a hundred m length or more as described in the previous chapter, the prototyping and development of devices were started. The target devices are from wide-ranging fields: power application equipment such as power cables, transformers, superconducting magnetic energy storage (SMES), and current limiters; medical application equipment such as magnetic resonance imaging (MRI), heavy particle accelerators, and nuclear magnetic resonance (NMR); and mobile equipment applications such as generators and motors.

In these applications, the specifications expected of the tapes are not necessarily uniform due to the difference in operating environments of the respective equipment or devices. In the development described in the previous chapter, the in-field I_c served as an index of properties, but the capacity of the aforementioned equipment is an additional needed function. For example, in an application that utilizes magnetic fields such as motors, generators, MRI, and accelerators, high I_c in magnetic fields is necessary, and in many cases, it is also necessary to have properties that can withstand the mechanical stress generated by the magnetic fields. In the application using currents such as cables and transformers, the reduction of AC loss is the issue. AC loss is the loss incurred due to the interlinkage flux that shifts when AC is applied to the superconductor, and is dependent on the J_c and the transfer distance of the magnetic flux (width of tape). This property is the same in the armature coil of motors, and moreover, it is similar to the measures to control the effect of currents of shielded MRI coils. The following is the description of the technology to improve the I_c (J_c) properties in magnetic fields and the development of a low-loss tape.

It is known that the installation of artificial pinning centers can improve J_c properties in magnetic fields. In capturing the quantized magnetic flux, it is necessary to disperse fine non-superconducting phase with the same order as the coherence length. Specifically, the dispersal of artificial pins at nm order is necessary in the REBCO material. For REBCO thin film tapes, development was started for the technology to install artificial pins mainly by a gas phase method. Various materials were targeted such as Y_2BaCu_5,[25] $BaZrO_3$,[26][27] and Y_2O_3.[28] However, good results were obtained with $BaMO_3$ (M = Zr, Ce, etc.) to which artificial pins (nano rods) oriented to the c-axis of REBCO at nano-size were introduced. Originally, REBCO materials possess large anisotropy reflecting their structure. In the applied magnetic field angle dependence of J_c properties, the J_c

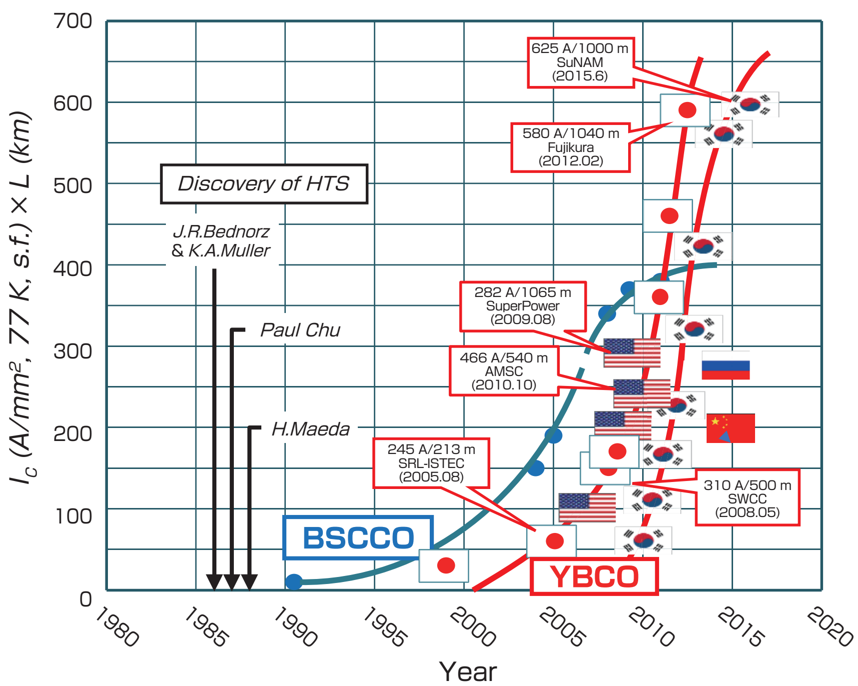

Fig. 9 Conceptual diagram of IBAD method[24]

(B//c) value, which is the J_c property in an environment in which an external magnetic field is applied parallel to the c-axis of superconducting crystal orientation, shows significantly lower behavior against the J_c (B//ab), which is the J_c property in an environment in which an external magnetic field is applied parallel to the a-axis or b-axis of superconducting crystal orientation. Therefore, by installing the aforementioned nano-rods, we obtained improvement of J_c (B//c) that has essentially a low J_c property.[25][26] In this field, the issue of development was how to disperse fine non-superconducting phase while maintaining high crystallization, or the technology to control the structure. To solve this issue, we recently found $BaHfO_3$ as an effective artificial pin material.[29] It became possible to form tapes that maintained the J_c (B) properties even to thick film regions by combining EuBCO as a superconducting layer, and the I_c properties were achieved in large magnetic fields (Fig. 10).[30][31] In this combination, since partial melt growth occurred, it was confirmed that the length and dispersal of $BaHfO_3$ do not depend on thickness, while achieving high crystallization, and this is thought to be the reason high J_c (B) properties are maintained in thick superconducting film layers. The achievement of long length is being investigated using this combination, and Fig. 11 shows the achievement of almost the same properties as short-length samples with thick films (3.5 µm) of EuBCO + BHO. Moreover, Fujikura reported high properties for tapes at a manufacturing level utilizing the above combination.

On the other hand, in the TFA-MOD method that is highly expected as a low-cost process, we engaged in the development of an installation process of artificial pins similarly to the aforementioned gas phase method. The representative material has been confirmed to be $BaZrO_3$, just as in the gas phase method.[32] However, the form is spherical (nanoparticles) and this is completely different from the nano-rods of the gas phase method. This arises from the growth mechanism of the film. A gas phase method like PLD is a system in which deposition seeds are supplied to a growth interface from a target, and superconducting layers and artificial pinning centers grow in almost the same interface, while the TFA-MOD method is a system in which a calcinated film that is made by calcining a coated film at low temperature is present on an intermediate layer, and artificial pin materials nucleate and grow there before a superconducting layer is formed, and there is no orientation relationship with the intermediate layer. Later, a superconducting layer grows from the intermediate layer as nanoparticle artificial pins that are dispersed in the front are incorporated, as a superconducting layer undergoes epitaxial growth. Therefore, nanoparticles that are random and have no orientation relationship are dispersed inside the oriented superconducting layer (Fig. 12). Reflecting the difference in the form, an equivalent improvement effect is seen against the applied orientation of the magnetic field. However, it was difficult to miniaturize the pins further, and the values were lower than the J_c (B) of the gas phase method. To solve this problem, we first developed miniaturizing technology by optimizing the heat treatment condition.[33] In this method, it is thought that the artificial pinning centers can be nucleated and grown at low temperature by heat treatment over a certain time at medium temperature of the intermediate heat treatment, or somewhere between calcination and full-firing, to complete the necessary phase change beforehand. The effect is the achievement of small artificial pinning centers with stunted growth. More recently, we succeeded in dramatic miniaturization by reducing the coating film to ultrathin (150 → 30 nm).[34][35] This is called the ultra-thin once coating (UTOC) MOD method, and greatly improves the in-field properties of the MOD method. Moreover, by changing the artificial pin material from $BaZrO_3$ to $BaHfO_3$ and increasing the added amount (10 → 25 mol%), we achieved 4 MA/cm^2 (65 K, 3 T) that surpassed the in-field J_c properties for tapes in the gas phase method.[36]

It was known that it was possible to reduce AC loss by creating filaments. In metal superconductors, low-loss is achieved by bundling and twisting ultra-thin wire, but it is difficult to handle ultra-thin wire created from tape material with a layered structure, and it was necessary to develop different technology. To solve this issue, first, we developed

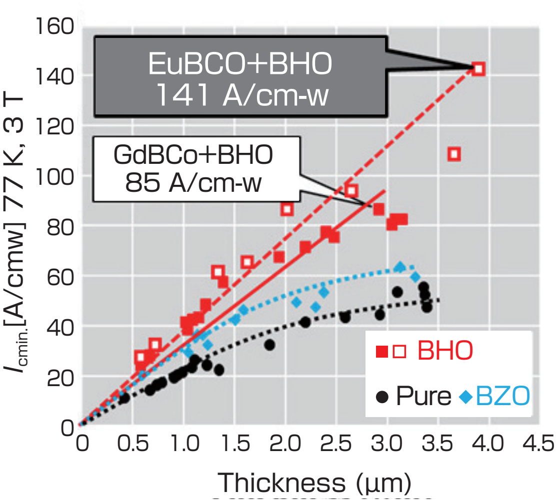

Fig. 10 Effect of artificial pinning center material against film thickness dependency of in-field I_c property for PLD film[31]

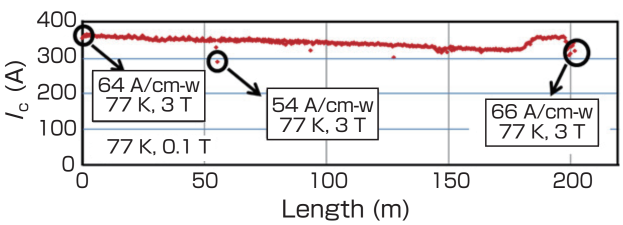

Fig. 11 PLD long-length tape with artificial pinning center[31]

scribing technology in which the superconducting layer would be thinned without separating it from the substrate. There were several different methods proposed around the world, and we developed a method in which a resin tape is placed on the surface of a tape, only the area where the groove will be made is melted with a heat laser, and then conducting chemical etching.[37] This method enables long-length scribing, and we achieved division processing of a 100 m class (Fig. 13).[31][38] However, this method had issues such as the groove width was uneven, and peeling occurred if there were any over-etched areas. Therefore we developed fine scribing technology using excimer lasers without using liquid.[39] This method allowed the fabrication of narrower width (100 → 30 μm) as well as of even groove width. Importantly, the scribed tape achieved AC loss reduction at the tape level, but this effect is often lost in the coil form that is necessary in application. The loss reduction effect was successfully retained even in the coil form through special coiling technology. It has been applied to transformers and armatures of full superconducting motors, and the principle verifications are being conducted.[40][41]

5 Future prospects

As mentioned above, it was initially difficult to achieve tapes with high properties and long length due to the difficulty of handling REBCO superconducting tapes, but dramatic progress was seen due to achievement of highly oriented intermediate structures, development of technology to form superconducting layers, installation of artificial pinning centers, development of fine wire technology, and others. Since then, development is being done for power cables, transformers, limiters, SMES, marine motors, MRI, accelerators, and others. For all these devices, there has been progress such as fabrication of instruments using superconducting tape and confirming a certain degree of performance, but currently there is no equipment that has been put to practical use.

There are three main reasons for this. First is the high cost of tapes. The REBCO tape tends to have high cost due to a complex fabrication method, and although some cost reduction can be expected by reducing the amount of tape used through improved properties, there is a limit in the amount of current that can be passed through one tape from

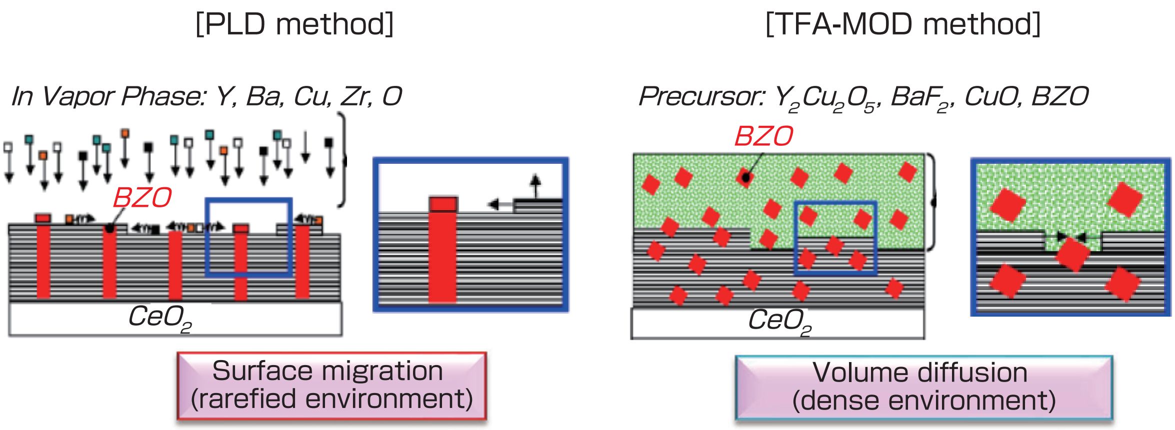

Fig. 12 Different growth mechanisms for films containing artificial pinning centers in PLD and TFA-MOD methods

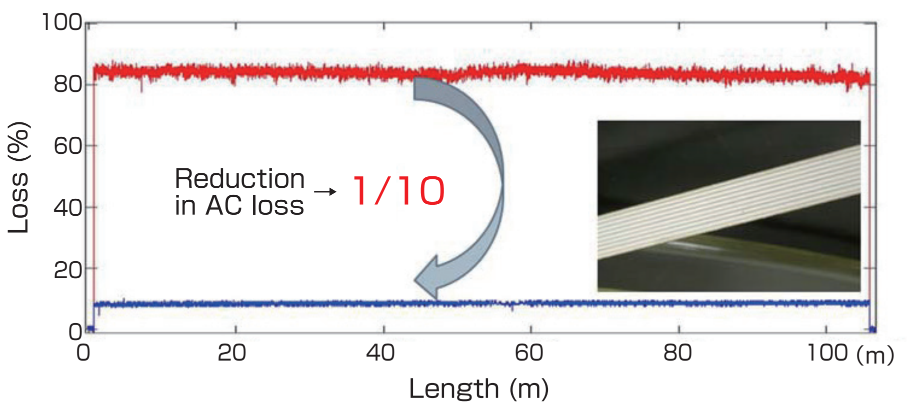

Fig. 13 Achievement of low AC loss in 100 m class tape material using scribing technology[31]

the perspective of energy recovery and protection. In general, 700−800 A/cm range is the limit as the operating current, and considering the load factor, 1300–1500 A/cm range is the maximum critical current. Therefore, if this critical current is not reached in the environment of operating temperature and magnetic field for each equipment, essential cost reduction can be achieved by improving the operating current. Currently, the above properties are already satisfied at 50 K or less with 1–2 T of external magnetic fields, but further development is needed for expected high temperature (e.g. liquid nitrogen temperature 65–77 K) and medium to high magnetic fields (3 T or more).

Another cause of high cost is the low yield. To truly increase the yield, it is necessary to improve the uniformity, but it is not easy to fabricate a narrow tape in km order without any areas of property degradation. Therefore, it is necessary to establish repair technology that enables recovery and increases stability.

The second reason why a superconducting device has not reached practical application is that there is no established overwhelming superiority against the existing technology. Although there is much equipment for which functions have been verified, a user will select the existing technology backed with past good results unless the new technology is absolute. There needs to be something that can only be realized with superconducting devices. For example, although there is no great superiority of a device itself, clear superiority may be seen when by using the superconducting device, a building can be kept very small and there is overwhelming cost merit in doing so. Such superiority is necessary in the initial introduction of this new technology.

The third important factor is the level of commitment of the end user. Of course, this links to the first two reasons, and along with overwhelming superiority, the user must be committed to participating in the development of a technology that will become necessary in the present or near future. The REBCO superconducting tape discussed in this paper has great attractiveness due to many advantages, but also has several disadvantages. A representative disadvantage is low handling due to it being a tape. Compared to wires, it lacks malleability and it is difficult to be arranged in coils or complex shapes. As a measure, the technology to form wire has been attempted, but this is not easy in terms of maintaining uniformity. Therefore, when conducting device development under certain limitations, it is necessary to consider the structure and operation that can be accomplished uniquely with a superconductor tape, rather than simply replacing metal wire.

Development is being conducted by many engineers around the world, but progress is slow at this point. The author thinks that one of the applications that may provide a breakthrough is the application to electric propulsion aircraft. To respond to the requirement for CO_2 reduction, the aviation industry is trying to shift from jet propulsion to electric propulsion. However, if one tries to achieve motorization by normal conductivity (iron and copper), the medium to large aircraft will become extremely heavy, and that is disadvantageous for aircraft. Therefore, an electric propulsion system using superconductor technology is expected. This is an idea of building a lightweight and high-output propulsion system using superconductors with full superconducting generators, motors, cables, and others. Since aircraft itself is expensive, the cost ratio of the tape will be small, and the tape cost may cease to be a problem. Since large aircraft is difficult unless superconductors are used, it is likely to be a major candidate if the user sets his mind on such development.

Overviewing the current situation, the high-temperature superconductivity technology is thought to be in the “valley of death” that is often encountered by new technology. To break out of this situation, it is necessary to achieve the first practical application by solving the aforementioned issues. For the superconductor technology to diffuse widely, it is important to show the effectiveness of this technology to the world through practical application.

Acknowledgement

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Graduated from the Graduate School of Engineering, Tohoku University in March 1987. Doctor (Engineering). Joined the Sumitomo Metal Industries, Ltd. and was dispatched to International Superconductivity Technology Center (ISTEC) during 1989–1993. Joined ISTEC in 1998, and then joined AIST in 2016 upon dissolution of ISTEC. Consistently engaged in the development of magnetic materials, oxide superconductor materials, and silicon monocrystal materials. Since 1998, engaged in the development of an RE superconductor tape process and its application, and was involved in many national projects. Acted as project leader. In this paper, described the development of RE superconductor tape material based on the experiences in these projects.

Discussions with Reviewers

1 Overall

Comment (Toshihiko Kanayama, AIST)

Ever since the discovery of high-temperature oxide superconductors in 1986, the development and practical application of wires and tapes using the material has been a technology of dreams that has not been realized, despite great expectations. This paper describes the issues and courses of development centering on the results obtained by the author, selection of materials and synthesis methods, practical fabrication technology for long-length tapes, and technology to improve properties. It is also mentioned that national project research involving companies was effective in this process. It is necessary to integrate diverse elemental technologies to link a discovery of a new substance to practical products, and this is a significant example that illustrates the necessity of continuous development over many years by a number of researchers.

Comment (Kei'ichi Ikegami, AIST)

This paper is about the development of a rare-earth superconducting tape material that is on the verge of practical realization, and that is expected for use in devices in the future. It explains the origin and the current situation comprehensively in an understandable manner, and it is well worth reading as a technological guidebook with a historical perspective.

2 R&D scenario

Comment 1 (Kei'ichi Ikegami)

The "scenario" on which *Synthesiology* places much value is set by a certain subject, while in this paper, to emphasize objectivity, the presence of a subject or agent, who set the scenario and pushed the development, has become weak. As a *Synthesiology* paper, the paper should focus as its subject of analysis not on the superconducting material, but on case studies of development. Therefore, I think you should make additions to clarify the core agent of the case studies.

You present Fig. 1 as your scenario, but when I read this paper, I see that the R&D was conducted by three different scenarios for three issues at stake. On the other hand, *Synthesiology* states: "To describe the elemental technology (technologies) that was (were) selected to realize the research goal. To describe the reasons for selecting the elemental technology (technologies). To describe in the terminology of science and technology how the selected elements are interrelated to each other, and how the elements were integrated and synthesized to realize the research goal." Therefore, in addition to Fig. 1, I think you should have a "figure for a detailed scenario" that shows how you selected the elemental technologies and how they were synthesized and integrated to realize the research goal.

Comment 2 (Toshihiko Kanayama)

This paper describes the important points of a general description of the chronology of development of oxide superconducting tapes. However, it is not clear how the author or the institution to which the author belongs made selections that led up to the development, that is the main focus of *Synthesiology.*

To clarify this point, how about adding a table of elemental technologies? I think the significance of this paper will be greatly enhanced if the reader can easily see what kinds of elemental technologies were candidates of the three issues shown in Fig. 1, and which ones the author or the institution selected.

Answer (Teruo Izumi)

I summarized the elemental technologies for each issue in Table 1.

Comment 3 (Toshihiko Kanayama)

The addition of Table 1 made the outline of the structure clearer. However, many of the elemental technologies remain as mere listings of technological issues that were targeted. For example, how about setting a third column with a title such as "Breakthrough technology" in correspondence to the elemental technologies of the second column? Then, I think the flow of the paper can be discerned easily if you describe the solutions that became keys to development of each elemental technology issue that is discussed in this paper.

Answer (Teruo Izumi)

As you indicated above, I revised the table.

3 Core agent of R&D

Comment 1 (Kei'ichi Ikegami)

Figure 1 is shown as the scenario, but who set this scenario? It seems to be shared among the worldwide research community which was involved in the development of rare-earth superconductors, and it also seems to be the strategy employed by the research group at ISTEC (AIST) to which the author belongs. I think you should clarify the main agent of the development that you are analyzing at the beginning of the paper.

Answer (Teruo Izumi)

The scenario in Fig. 1 can be called the strategy of ISTEC (AIST), and I added this fact to the text.

The main agent of the development was written clearly at the end of "1 Introduction."

Comment 2 (Toshihiko Kanayama)

Please write up separately the world trend of development and the items promoted by the author and the institution to which the author belongs.

Answer (Teruo Izumi)

To clarify the items conducted by ISTEC (AIST), the subject "ISTEC" or "we" were added to the text.

4 R&D organization and national project

Comment 1 (Kei'ichi Ikegami)

It can be seen that in the research analyzed in this paper, the organization called ISTEC seems to have played a very important role. Please describe how ISTEC came to be, its mission, positioning in the national strategy, leadership of this organization, how and which researchers were gathered, and how these researchers worked on R&D through their union, collaboration, and division of labor.

Comment 2 (Toshihiko Kanayama)

This paper mentions that the national project research was effective, but it does not mention the content or the organizational structure. Please add some simple explanation about ISTEC and the NEDO projects.

Answer (Teruo Izumi)

I added a description of ISTEC at the end of "1 Introduction."

5 International competition

Comment (Kei'ichi Ikegami)

I think there was fierce competition of development against overseas teams, especially the Americans, and I am curious how this affected the scenario setting and execution. Can you add some text about this?

Answer (Teruo Izumi)

For the effect of competition between Japan and USA as you mentioned, text was added to the end of "3 Second issue."

6 R&D issues and required values

Comment 1 (Toshihiko Kanayama)

The relationship between the achieved value and the required value for electric current that is described as the issue for practical realization in "Future prospects" is not clear. Please add some text on this matter. Also, I think the positioning of this paper will become clear if you add, if possible, the issues that remain toward practical realization and the candidate solutions.

Answer (Teruo Izumi)

I added a text to "Future prospects."

Comment 2 (Kei'ichi Ikegami)

In "2 First issue," you state, "obtaining a J_c property of about 10^3 A/cm^2 (77 K, self-generated magnetic field) was the best we could do." I don't think the readers will understand the meaning of these numbers unless you indicate other numbers for comparison, such as how many J_c properties are needed for practical realization, or what level of J_c properties has been realized in tapes of other materials.

Answer (Teruo Izumi)

I added explanations on the points indicated above.