STUDY OF MgB\(_2\) THIN FILMS FOR ACCELERATOR CAVITIES IN KEK

M. Fukutomi, K. Kawagishi, H. Kitaguchi, Y. Kobayashi, K. Komori, H. Kumakura, A. Matsumoto, National Institute for Material Science (NIMS), Tsukuba, Ibaraki, Japan

Abstract

Because of the higher transition temperature of MgB\(_2\) than that of Nb, the microwave surface resistance \(R_s\) of MgB\(_2\) could be expected to be much lower than that of Nb. Further, though the value of \(H_{c2}\) is almost comparable, the Ginzburg-Landau parameter, \(\kappa\), of MgB\(_2\) is almost fifty times larger than that of Nb. If MgB\(_2\) is applied to accelerator cavities, it leads to a conclusion that the theoretical limit of the electric field could be larger than that of Nb by more than 40\%. We describe the present status of our study of MgB\(_2\) thin films for accelerator application.

STUDY OF MICROWAVE PROPERTIES OF HIGH-\(T_C\) SUPERCONDUCTORS IN KEK

Soon after the discovery of high-\(T_C\) superconductors [2] in 1986, especially after the discovery of YBaCuO [3, 4], we have been interested in the application of these materials to accelerator cavities. We intend to make films on the surface of metallic materials, because of good thermal conductivity and the mechanical machinability. We started to fabricate films on a part of a cylindrical cavity, mainly on one of the endplates and once on the cylindrical part, and to measure the microwave properties in the TE\(_{011}\) mode. We can divide the research effort to four different stages. First we made YBaCuO thick films by a plasma-spray method on 3 GHz cylindrical cavities [5]. Second we made BiSrCaCuO thick films by a screen-printing or spray-coating method on 3 GHz cavities [10]. Third we fabricated YBaCuO thin films by a PLD method on 16 GHz cavities [7, 11, 13, 14]. At the present fourth stage, we make MgB\(_2\) [15] thin films by a PLD method on 13.6 GHz cavities. The substrate material are extensively copper except only one silver sample. In the first and the second stage, the diameter of the endplate was as large as 15 cm, and the crystal orientation was uncontrolled. We obtained around 0.2 m\(\Omega\) of the surface resistance for YBaCuO films on silver. In the third stage, the NIMS group of us developed a modified-bias-sputtering technique [6, 8] by which films could be grown so that the c-axis is normal to the surface. However, it was shown that \(R_s\) of these samples were dependent on rf power [11] as shown in Fig. 1 and the critical current density depends on the temperature [14] as shown in Fig. 2.

In this paper, we report fabrication of MgB\(_2\) films on copper by a precursor post-annealing process, and some of the results of the rf measurement.

Figure 1: Microwave field dependence of the surface resistance.

Figure 2: The critical current density vs. temperature. Sample EC403 and EC392 are well-textured, and EC391 and EC425 are weakly-textured.

REQUIREMENT FOR SUPERCONDUCTING CAVITIES

In accelerator cavities, low rf loss \(P_{\text{loss}}\) and high rf field level \(E_{\text{acc}}\) are two important factors to be considered. The former requires small microwave surface resistance, \(R_s\), since \(P_{\text{loss}} \propto R_s H^2\). The latter requires large superheating magnetic field, \(H_{\text{sh}}\), since \(E_{\text{acc}} \propto H_{\text{sh}}\). In these point

---

\(\text{*Work partially supported by Grant-in-Aid for Scientific Research (KAKENHI).}

\(\text{\textsuperscript{1}inagakis@post.kek.jp}\)
of view, MgB₂ films could be a good candidate for superconducting cavities.

**Microwave surface resistance**

The microwave surface resistance, \( R_s \), is the sum of a residual resistance \( R_{\text{res}} \) and a resistance \( R_{\text{BCS}} \), which depends on the frequency and the temperature.

\[
R_s = R_{\text{res}} + R_{\text{BCS}} = R_{\text{res}} + \frac{\Delta^2}{T} \exp\left(-\frac{\Delta}{k_B T_c} \cdot \frac{T_c}{T}\right),
\]

where \( \Delta \) is the energy gap, \( k_B \) the Boltzmann constant and \( T_c \) the critical temperature. After Eq. (3.30), \( 2\varepsilon_0/k_BT_c = 3.50 \), in the BCS paper [1], it is described that \( \text{from the law of corresponding states, this ratio is predicted to be the same for all superconductors.} \)

In the case of Nb, \( T_c = 9.23 \) K and \( \Delta = 1.40 \) meV leads to \( 2\varepsilon_0/k_BT_c = 3.52 \). In the case of YBCO, \( T_c = 78 \) K and \( \Delta = 11.40 \) meV leads to \( 2\varepsilon_0/k_BT_c = 3.02 \). In the case of MgB₂, \( T_c = 39 \) K and \( \Delta = 5 \) meV leads to \( 2\varepsilon_0/k_BT_c = 2.98 \). Considering all things together, if we take \( \Delta/k_BT_c = 1.75 \), we obtain Table 2 at \( T = 4 \) K.

<table>
<thead>
<tr>
<th>material</th>
<th>( T_c ) (K)</th>
<th>( \exp(-\alpha T_c/T) )</th>
</tr>
</thead>
<tbody>
<tr>
<td>Nb</td>
<td>9.23</td>
<td>0.015</td>
</tr>
<tr>
<td>Nb₃Sn</td>
<td>18.3</td>
<td>4.9 \times 10^{-4}</td>
</tr>
<tr>
<td>YBa₂Cu₄O₇</td>
<td>92</td>
<td>2.25 \times 10^{-17}</td>
</tr>
<tr>
<td>MgB₂</td>
<td>39</td>
<td>8.7 \times 10^{-8}</td>
</tr>
</tbody>
</table>

It is clear that because of the low attribute to the exponential factor, MgB₂ cavities can be adopted even at higher frequencies compared with Nb, which is now used at c-band frequency at an evacuated temperature of liquid helium.

In passing, reducing \( R_{\text{res}} \) is another problem to be investigated.

**Superheating field**

In the case of dc, the figure of merit of the critical field is \( H_{c1} \) or \( H_{c2} \). Meanwhile, in the case of rf, the figure of merit is the superheating field \( H_{sh} \). Using the thermodynamic critical field, \( H_c \),

\[
H_{sh} \simeq \frac{H_c}{\sqrt{\kappa}}, \tag{2}
\]

where \( H_c = H_{c2}/\sqrt{\kappa} \) and \( \kappa \) is the Ginzburg-Landau parameter [12]. Since the theoretical limit of the electric field in an accelerator cavity, \( E_{\text{acc}} \), is related with \( H_{sh} \) as

\[
H_{sh}/E_{\text{acc}} = 420e/\text{MV/m} = 3.34 \times 10^{-3} \text{A/m/V/m}, \tag{3}
\]

we have Table 1.

This shows that \( E_{\text{acc}} \) of MgB₂ could be higher than that of Nb by 40%. This becomes a strong motivation to pursue accelerator cavities made of MgB₂. (Recently, one of the authors (K. Saito) showed that \( H_{sh} \propto H_c/\kappa \) rather than Eq. 2. Therefore, the values of Table 1 should be referred in a relative comparison.)

**Machinability-or how to make cavities**

The next to be considered is how to make accelerator cavities. One possible way is to make cavities of bulk materials. The hot isostatic press (HIP) method and the recently proposed reactive liquid Mg infiltration method [17] belong to this category. Another way is to make cavities of metallic material covered with superconducting films. We follow one of the latter way, i.e. a pulsed laser deposition (PLD) and heat treatment method, called precursor post-annealing method.

**PRECURSOR POST-ANNEALING METHOD**

This method consists of two steps:

- Fabrication of precursor by PLD
  - The target is a pellet with a mixture of MgB₂ and Mg powder with the stoichiometry of Mg:B=2:1. Excimer laser with \( \lambda = 248 \) nm (KrF), 400 mJ, and 5–10 Hz is irradiated on the target in \( 6 \times 10^{-5} \) Torr Ar atmosphere at room temperature for 2 hr. The thickness of the fabricated film is around 1 \( \mu \)m. The substrate material are either silicon, sapphire or copper (YSZ/Cr/Cu).
  - Post-annealing
    - Typically, the precursors are heated in 1 atm Ar gas at 550–650 °C for 10 min. This process must be optimized so that magnesium evaporation should be well controlled to become MgB₂ in the phase diagram [16], and to make the oxidation of magnesium as minimum as possible.

The reader is asked to refer the papers by Fukutomi et al. [6, 8] for further details.

**ASSESSMENT OF FABRICATED FILMS**

**Composition analysis**

In order to find the condition of PLD and post-annealing, we made films on silicon or sapphire strips of the size of around \( 12.5 \times 6.25 \times 0.5 \) mm. Composition analysis by inductively coupled plasma (ICP) showed that one of precursor samples contained 8.57 ppm Mg and 1.83 ppm B, from which we obtained the ratio Mg:B=4.16:2 as expected.

**Structure analysis**

Thereafter some samples were structure-analyzed by small angle X-ray scattering in the KEK photon factory.
Table 1: Theoretical limit of the electric field in an accelerating cavity.

<table>
<thead>
<tr>
<th>material</th>
<th>$T_c$ (K)</th>
<th>$\kappa$</th>
<th>$H_{c2}$ (A/m)</th>
<th>$H_c$ (A/m)</th>
<th>$H_{sh}$ (A/m)</th>
<th>$E_{acc}$ (MeV/m)</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pb</td>
<td>7.2</td>
<td>0.5</td>
<td>$1.9 \times 10^5$</td>
<td>$8.4 \times 10^2$</td>
<td>$6.4 \times 10^4$</td>
<td>20.1</td>
</tr>
<tr>
<td>Nb</td>
<td>9.2</td>
<td>0.78</td>
<td>$1.6 \times 10^5$</td>
<td>$8.4 \times 10^2$</td>
<td>$6.4 \times 10^4$</td>
<td>50.3</td>
</tr>
<tr>
<td>Nb$_3$Sn</td>
<td>18.2</td>
<td>22.8</td>
<td>$2.5 \times 10^4$</td>
<td>$6 \times 10^5$</td>
<td>$6 \times 10^5$</td>
<td>95</td>
</tr>
<tr>
<td>YBaCuO</td>
<td>94</td>
<td>700--1000</td>
<td>$1.8 \times 10^7$</td>
<td>$3.5 \times 10^5$</td>
<td>$2.6 \times 10^5$</td>
<td>25.2</td>
</tr>
<tr>
<td>MgB$_2$</td>
<td>39</td>
<td>36.3</td>
<td>$1.8 \times 10^7$</td>
<td>$3.5 \times 10^5$</td>
<td>$2.6 \times 10^5$</td>
<td>69.2</td>
</tr>
</tbody>
</table>

Figure 3 shows a relative intensity vs. angle, where violet and green lines are derived from the same precursor before and after annealing, respectively. The blue lines are attributed to silver, pasted on the back side of the substrate.

![Figure 3: Relative intensity vs. angle taken by a small angle X-ray scattering method in PF.](image)

**Measurement of the dc resistance**

The dc resistance was measured by the four terminal method. Figure 4 is the ratio of the resistance $R(T)$ at $T$ to that at 40 K for different post-annealed samples of the same precursor on sapphire strips. The onset transition temperature of the most samples were 28–30 K with $\Delta T_c \approx 2.5$ K except one sample, whose resistance decreased from above 40 K with a wide $\Delta T_c = 15$ K. We need further study to obtain qualified samples.

![Figure 4: Normalized resistance of differently annealed samples of the same precursor.](image)

**Principle to measure microwave surface resistance**

The microwave surface resistance was measured by the conventional host cavity method as shown in Fig. 5. The cavity comprises two components: one is machined out of a block of copper, the inner surface of which is of cylindrical shape with a flat bottom surface; the other is a top endplate. We call the former a host-cavity and the latter an endplate. The endplate is replaceable with other endplates covered with MgB$_2$ film.

At temperature $T$, if the unloaded quality factor of the cavity with a copper endplate is $Q_{0,Cu}(T)$ and that with a MgB$_2$ endplate is $Q_{0,Cu+MgB_2}(T)$, $R_{s,MgB_2}(T)$ is calculated by[5]

$$
\frac{R_{s,MgB_2}(T)}{R_{s,Cu}(T)} = k \left( \frac{Q_{0,Cu}(T)}{Q_{0,Cu+MgB_2}(T)} - 1 \right) + 1,
$$

(4)

![Figure 5: Copper host cavity excited at 13.6 GHz in TE$_{011}$ mode.](image)
In the right side of Eq. 4, the $Q_{0,Cu}$ must be used at the same temperature $T$ as the $Q_{0,Cu+MgB_2}$. At the beginning of this study, we found the coefficients of the polynomials of degree 9 in the KaleidaGraph fitting for almost 3000 measured $Q_{0,Cu}$-values as shown in Fig. 6, and then calculated $Q_{0,Cu}$ at $T$ using these coefficients. However, as typically shown in Fig. 7 at low temperature region and in Fig. 8 at high temperature region, the fitting (denoted by a red solid line) deviated from the measured points systematically. Figure 9 shows the difference between the measured value $Q_m$ and the fitted polynomial of degree $Q_{c(9pol)}$. There, at least clear eight nodes can be observed.

Therefore, instead of using the polynomial fitting, we

$$Q_{0,Cu}$$

where $k$, a geometrical factor, is defined by

$$k = \frac{\int H_0^2 \, dS}{\int_{Stot} H_0^2 \, dS}. \quad (5)$$

Here $H_0$ is the magnetic field parallel to the surface, and the integration in the numerator is over the whole surface of the cavity and that in the denominator over the surface covered with the MgB$_2$ film.

For a cavity with radius $A$ and length $L$ excited in the TE$_{nm}$ mode, $k$ is related with geometrical resistances $Z_T$ by $k = Z_{T}^{end}/Z_{T}^{tot}$, with

$$Z_{T}^{end} = \frac{Z_0}{2} \frac{\left( p'_{nm} \right)^2 + \left( \pi A/L \right)^2 \left( \pi A/L \right)^2}{\left( A/L \right) \left( \pi A/L \right)^2} \quad (6)$$

and

$$Z_{T}^{tot} = \frac{Z_0}{2} \frac{C_n}{C_{d1} + C_{d2}}, \quad (7)$$

where

$$C_{d1} = \left( p'_{nm} \right)^2 \left[ 1 + \left( n/p'_{nm} \right)^2 \left( \pi A/L \right)^2 \right], \quad (8)$$

$$C_{d2} = 2 \left( A/L \right) \left( \pi A/L \right)^2 \left[ 1 - \left( n/p'_{nm} \right)^2 \right], \quad (9)$$

and

$$C_n = \left[ \left( p'_{nm} \right)^2 + \left( \pi A/L \right)^2 \right]^{3/2} \left[ 1 - \left( n/p'_{nm} \right)^2 \right]. \quad (10)$$

In the above equations, $p'_{nm}$ is the $n$th root of $J_0(x) = 0$, and $Z_0 = \sqrt{\mu/\varepsilon}$ is the vacuum impedance. The dimension of the host cavity is determined so that the contribution from the endplate is as large as possible in practical constraint.

In the present experiment, the diameter and the length of the cavity are 33 mm and 19 mm, respectively. The endplate is a circular disk of 36 mm diameter and 3 mm thickness. The resonant frequency of the TE$_{011}$ mode is about 13.6 GHz and $k = 4.2714$. 

Figure 6: Measured data of $Q_{0,Cu}$

Figure 7: Measured Q-value and a fitted polynomial of 9th degree below 50 K.

Figure 8: Measured Q-value and a fitted polynomial of 9th degree above 250 K.
adopt the Gauss-Hermit interpolation. Figure 10 shows the differences between $Q_m$ and the Gauss-Hermit interpolation $Q_c^{(GH)}$ with $N=80$ and $\sigma = 0.5$ [9]. The differences are evenly scattered around zero and obviously decreased the systematic error as can be compared in both figures. This process eliminated an artificial interpolation especially at low temperature region and could be also useful in the assessment of the microwave surface reactance.

**Measurement of the MgB$_2$ microwave surface resistance**

Before a fabrication process to make films on copper was established, we made films on silicon or sapphire substrate, which were 40 mm square with the thickness of 0.5 mm. Figure 11 is a host cavity to measure the microwave surface resistance of this shape. The dimension of the cavity is same as shown in Fig. 5. The black square sample on the right component is a YBCO film on sapphire with CeO$_2$ buffer layer. We may report the measured results using this cavity elsewhere.

![Copper host cavity for endplate of thin rectangular shape](image1)

Figure 11: Copper host cavity for endplate of thin rectangular shape

Figure 12 is endplates with MgB$_2$ films on copper substrate, and the measured value of the best sample obtained at present is shown in Fig. 13. The temperature controller was very unstable and the data were very scattered in this measurement. Anyway the left ordinate is the $Q_{0, Cu + MgB_2}$ denoted with thick dotted line and $Q_{0, Cu}$ denoted with thin dotted line. The right ordinate is the $R_{s, MgB_2}$ denoted with thick solid line and $R_{s, Cu}$ denoted with thin solid line. We fitted $R_{s, MgB_2}$ using Eq. 1 with $T_c = 27.0$ K, and obtain $R_s = 0.862$ m$\Omega$ and $\Delta/k_B T_c = 5.2816$. This is about three times of 1.76 described in the subsequent sentences.

![Copper endplate covered with MgB$_2$ films](image2)

Figure 12: Copper endplate covered with MgB$_2$ films.
of Eq. 1, meaning that $\Delta$ is much larger because of lower $T_c$. These discrepancies can be attributed to the film quality, and especially to the oxidation of magnesium.

![Figure 13: Microwave surface resistance of a MgB$_2$ end-plate denoted with a solid black line and a fitting with the BCS theory denoted with a red line.](image)

**FABRICATION OF MgB$_2$ FILMS ON ACCELERATING CAVITIES**

In the above description, we have been concentrated on how to make good films on metallic surfaces. Therefore the microwave surface resistances were measured using a TE$_{011}$ mode cavity, so that the current does not flow through the contact plane between the cavity components. However, in order to make accelerator cavities, the TM-like field need to be excited. To avoid interception of the current flow in the longitudinal direction, a cavity could be constructed by multiple components divided in the longitudinal direction, for example, as shown in Fig. 14 as a quadrant. However, since real cavities are complicated in shape, it is easier to cut cavities vertically to the axis. In this case, especially in superconducting device, making good contact between component-planes is a serious task. At present, we are making a cavity, which is a 1/3 scaled model of the ILC superconducting cavity. (The original frequency is 1389.63 MHz, and $Q_0 = 3.01 \times 10^5$ for copper at room temperature and $9.51 \times 10^9$ for niobium at 2.7 K.) The cavity consists of two parts made of OFHC copper as shown in Fig. 15. Before machining, they are annealed. The contact plane is lathed with a diamond bit to a flat mirror-plane as shown in Fig. 16. Since the mirror plane is so complete that it is uneasy to detach, once these two components come into contact. This technique is derived from the experience of the X-band linear collider project. The contact plane around the inner surface is chamfered with 0.1 $R$ as shown in Fig. 17 to avoid swelling inside the cavity surface. The inner surface of the cavity is also mirror-finished. After assembling, the inner surface will be covered with a precursor by PLD and post-annealed.

![Figure 14: A quadrant of a TM mode cavity.](image)

**SUMMARY**

After a long interruption of working condition, we are now able to make films by a precursor post-annealing method. Especially our effort will be directed toward making qualified films on copper substrate. And even the transition temperature is not a maximum as is should be, we try to develop a fabrication technique to be usable for accelerator-mode cavities.
ACKNOWLEDGEMENTS

We would like to express our thanks to Dr. Eiji Takayama-Muromachi and Dr. Kiyoshi Inoue, the former director of Superconducting Material Center in NIMS for giving the convenience to use facilities of NIMS. Dr. Kazumasa Togano is acknowledged for giving the advice for this study. Dr. Yuichi Morozumi is acknowledged for giving information of the ILC superconducting linear collider. We express our thanks to Messrs. Satoru Nakayama and Tomio Kubo for their help in the experiments.

REFERENCES


