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Replacement of NbN by $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ in superconducting thin film coil in a spiral trench on a Si-wafer for compact SMESs

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Abstract. To improve the energy storage volume density of our concept of a compact superconducting magnetic energy storage system, replacement of superconducting material NbN for the thin film coil in the spiral trench on a Si wafer by a high temperature superconductor $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ was addressed. By using the successful result previously reported to grow a c-axis oriented film on a steel substrate, a superconducting film of $T_c=45.5$ K was obtained using similar buffer layers including a (100) oriented CeO_2 layer over a (100) oriented YSZ layer. The similar result reported on a SrTiO_3 single crystal substrate without any buffer layers indicates c-axis oriented film of $T_c=90$ K can be obtained by further reduction of oxygen partial pressure.

1. Introduction

We have been developing superconducting thin film coils in a spiral trench on a Si wafers for compact SMESs as shown in figure 1. A proof of concept has been performed using NbN thin films showing energy storage of 0.01 mJ [1]. Increasing NbN thickness by mitigating film stress [2], the stored energy increased up to 0.1 mJ [3]. Figure 2 shows a Si wafer of 101.6 mm in diameter with a latest design of the spiral trench. The 36 sided-polygonal trench in figure 1 has been changed to the 144 sided-polygonal trench. Parallel trenches are connected each other by occasional interconnections to form a bundle of 7 trenches. This bundle forms a spiral pattern throughout the wafer. In this latest design, the sevenfold increase of the critical superconducting current I can be expected while the number of the turn of the spiral: n decreases. The inductance L decreases in proportion to n . This change is advantageous since the stored electric power is in proportion to $L \times I^2$.

For further improvement, we moved on to replacement of NbN by $\text{YBa}_2\text{Cu}_3\text{O}_{7-\delta}$ (Y123). This replacement is to enable use of a cold source of liquid hydrogen: 20K or even of liquid nitrogen: 77K instead of use of a cryogenic refrigerator at 13K or lower in the case of NbN. Another benefit of this replacement is that the critical superconducting current density of Y123 can be about 2×10^4 A/mm² which is about 18 times larger than that of NbN [4]. Since the amount of magnetic energy storage is proportional to the square of the current, we may expect $18 \times 18 = 324$ times larger energy storage in the case of Y123 in comparison to that in the case of NbN. Here, it should be noted that the



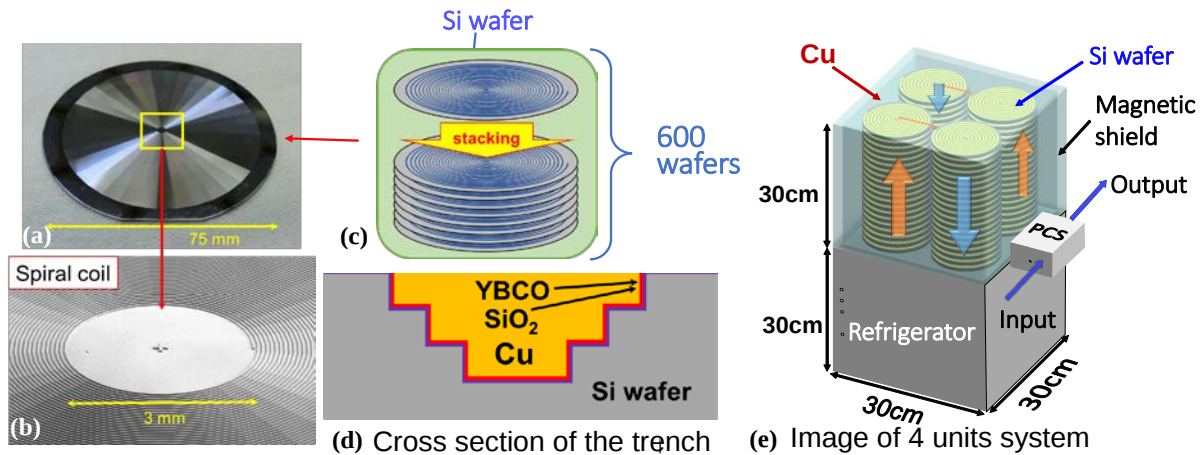


Figure 1. Superconducting coil composed of 4 coil-stack units. Each of the units is made of stacked Si wafers engraved with a superconducting thin film coil in a spiral trench formed with MEMS technology.

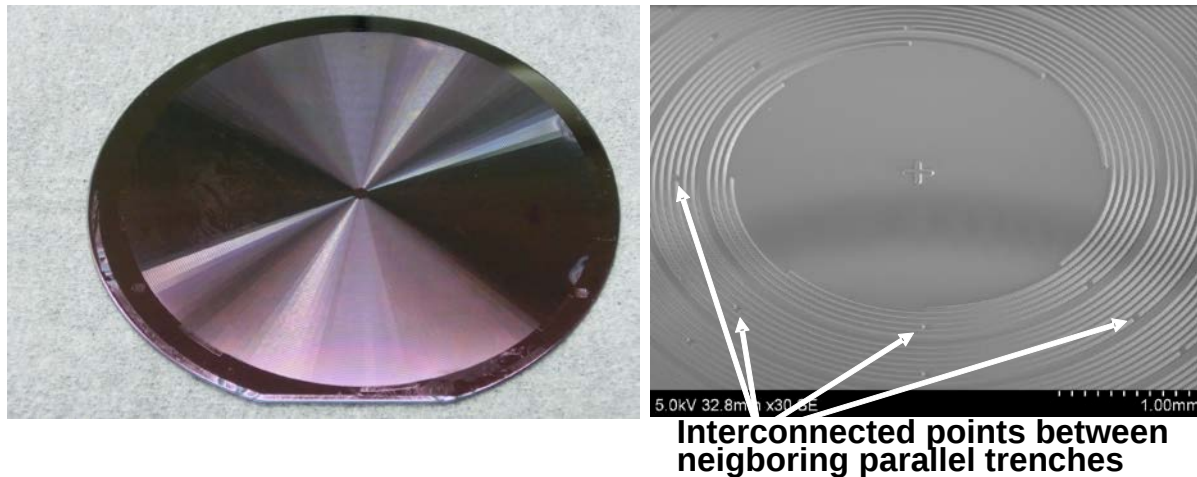


Figure 2. 144-sided polygonal spiral trench pattern of 7 parallel trenches connected each other by occasional interconnections on a Si wafer of 101.6 mm in diameter.

electromagnetic stress imposed on the superconducting coil becomes also 324 times larger. In a typical favourable design of the spiral coil in the case of NbN, the four units system shown in figure 1 together with a cryogenic refrigerator, gives the approximate estimate of the magnetic energy storage volume density as high as 1.8Wh/L and the highest electromagnetic stress near the central axis of the coil unit this is as high as 0.11 GPa which is well below the mechanical strength limit of a single crystalline Si: 4 GPa. However, these values are estimated to be 583 Wh/L and 35.6 GPa, respectively, by the replacement of NbN by Y123. This means the maximum superconducting current in the coil must be much reduced from the value derived from the critical current density. We are doing a more precise estimation at present and will be reported elsewhere. Another problem of this replacement comes from the Cu-plating process which is done in the acidic solution in the case of NbN. Since this conventional acidic solution corrodes Y123 films, we performed extensive survey of the compositions of solution and Cu-plating conditions. However, we are still on the way to find an acceptable process. Here we came across a workaround method to form a sputter-deposited thin Cu film on the Y123 film before the conventional Cu-plating. By covering the Y123 film with a sputter-deposited Cu film including the edge portion of the wafer before Cu-plating, we found the conventional Cu-plating was successfully used as shown in figure 3.

2. Experimental

Since we utilize sufficiently matured Si-MEMS technology to form the spiral trenches as shown in figure 2 as well as the mechanical strength of the single crystal Si wafer, superconducting Y123 films must be formed on Si wafer for the present purpose. Since the superconducting current increases with the cross-sectional area of the Y123 film in the trench, Y123 films should be deposited as thick as possible. For this purpose, fluorine free metalorganic deposition (FF-MOD) by dipping process is preferable. To form Y123 films by FF-MOD, most of the works have utilized epitaxial growth on single crystal substrates, such as on SrTiO_3 (STO) (100) substrate [5],[6], on LaAlO_3 (001) substrate [7], on CeO_2 -buffered R-plane sapphire (-1012) substrate [8-11], and on 40-nm-thick CeO_2 -buffered yttria-stabilized zirconia (YSZ) (100) substrate [12],[13]. In most cases, acetylacetonates ($\text{Y}:\text{Ba}:\text{Cu}=1:2:3$) based solutions were used. According to the previous

work [5], for example, c-axis oriented Y123 films show metallic temperature dependence of resistivity in the normal state and have narrow superconductive transition widths around 90K, while the films which have both c-axis and a-axis orientation show semiconductive behaviour in the normal state and show rather broad transition around 57K. Therefore, formation of c-axis oriented Y123 films (c-Y123) on Si substrate is preferable in our purpose.

On Si substrate, however, few works by MOD have been reported, but some have been reported by sputter-deposition [14], [15]. In these studies, a (100) oriented YSZ layer was firstly sputter-deposited on a Si (100) substrate succeeded by sputter-deposition of a (100) oriented CeO_2 layer (CeO_2/YSZ buffered layer) before the sputter-deposition of Y123 layer which turned out to be c-axis orientation. Therefore, we decided to form a thin Y123 layer by sputter deposition on the sputter-deposited CeO_2/YSZ buffered layer on a Si(100) wafer before the main Y123 layer formation by FF-MOD in the hope that this thin sputter-deposited Y123 layer may help the succeeding growth of c-axis Y123 film by FF-MOD.

More recently, c-Y123 layer was successfully formed on a CeO_2/YSZ buffer layer on Y_2O_3 coated steel substrates using propionates ($\text{Y}:\text{Ba}:\text{Cu}=1:2.1:3.15$) with additional HCl 0.1 [16-18]. In these cases, the initial precipitation of $\text{Ba}_2\text{Cu}_3\text{O}_4\text{Cl}_2$ crystallites on the CeO_2 surface was considered to be preferable for succeeding c-Y123 growth. Since this c-Y123 layer has no epitaxial relation to the steel substrate, this buffer-layer system may work also on a Si substrate for c-axis orientation of Y123.

Taking these previous results into account, we tried to form FF-MOD Y123 layer on a thin Y123 layer sputter-deposited on CeO_2/YSZ buffered Si (100) layer. However we experienced that addition of HCl into propionates solution was apt to cause unwanted sedimentations and we were not successful to make stable solutions for MOD. Therefore, we added dichloroacetic acid (DCA) into propionates ($\text{Y}:\text{Ba}:\text{Cu}=1:2.1:3.15$) solution in place of the HCl addition. For comparison, we also tested the commercially available acetates solution for MOD: Kojundo Chemical Lab., YBC-05(1/2/3).



Figure 3. Homogeneously Cu-plated Y123 film on a Si substrate of 101.6 mm in diameter.

3. Results

3.1. YSZ and CeO₂ sputter deposition on Si wafers

Firstly, sputter-deposition of YSZ on Si (100) wafers succeeded by CeO₂ has been performed to form CeO₂/YSZ buffer layer. Secondly, sputter-deposition of Y123 was performed on the CeO₂/YSZ buffer layer. Table 1 summarizes the process conditions of YSZ and CeO₂ sputter-deposition on Si wafers.

Table 1. Process conditions of each layer.

	Gas flow rate (sccm)	Gas pressure (Pa)	Substrate temperature (°C)	process time (h)
YSZ-deposition (500 nm)	Ar:50	5.0	575	3
CeO ₂ -deposition (500 nm)	Ar:50	5.0	575	3
YBCO Deposition (100nm)	Ar:2, O ₂ :10	24	670	3
Annealing	O ₂	9.0×10^{-4}	450	>1

Figure 4 shows X-ray diffraction patterns after YSZ deposition and after CeO₂ deposition. After YSZ deposition, a clear YSZ (200) peak can be observed. After CeO₂ deposition, a broad peak appeared overlapping the Si (200) peak which appeared the diffraction pattern after YSZ deposition.

This broadness of the peak teaches this peak is not of Si (200) peak originated from the single crystal Si (100) substrate but of CeO₂ (200) peak originated from the CeO₂/YSZ buffer layer.

3.2. YBCO deposition by FF-MOD

Deposition of Y123 was performed by FF-MOD using the acetate solution and the propionate solution at various annealing temperatures such as 713 °C, 733 °C, 753 °C ...etc. and O₂ partial pressure of 10 Pa (N₂ balance) for 75 min. At higher temperature such as 830 °C, formation of remarkable BaCeO₃ was observed. Figure

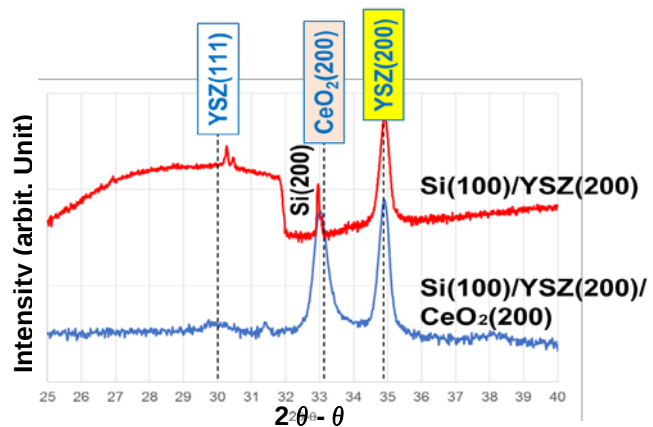


Figure 4. X-ray diffraction patterns after YSZ deposition (upper) and after CeO₂ deposition (lower).

5 displays the schematic cross-sectional structures of the three samples. X-ray diffraction peaks of Y123 are scarcely observed in sputter-deposited Y123. Figure 6 shows X-ray diffraction patterns of FF-MOD samples (a) by acetate solution, (b) propionate solution with DCA and (c) propionate

	Sputtered film	Acetate-MOD solution*	Propionate-MOD solution*
Cross section	Sp-YBCO 100nm CeO₂(100) 500nm YSZ(100) 500nm Si wafer	MOD-YBCO 200nm Sp-YBCO 100nm CeO₂(100) 500nm YSZ(100) 500nm Si wafer	$T_c = 45.5K$ MOD-YBCO 200nm Sp-YBCO 100nm CeO₂(100) 500nm YSZ(100) 500nm Si wafer
			(c/a-axis orientation)

Figure 5. Schematic cross-sectional structures of the samples.

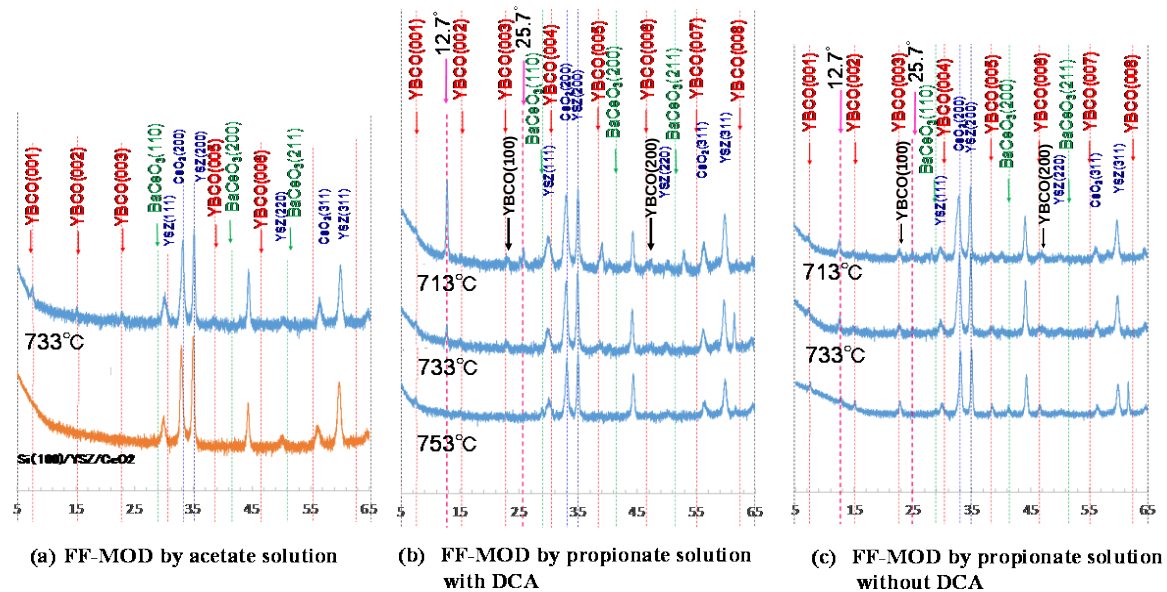


Figure 6. X-ray diffraction patterns of FF-MOD samples (a) by acetate solution, (b) propionate solution with DCA and (c) propionate solution without DCA.

solution without DCA. Diffraction peaks of Y123 are evidently observed in (a) FF-MOD by acetate solution. In the case of FF-MOD by propionate solution with DCA(b), c-Y123 and a-Y123 peaks can be clearly observed. The peaks at the diffraction angles of 12.7° and 25.7° corresponds to $\text{Ba}_2\text{Cu}_3\text{O}_4\text{Cl}_2$ crystallites as described in the previous reports [16–18]. However, the similar peaks were also observed in the diffraction patterns of the sample formed without DCA as shown in figure 6(c). These peaks cannot be of $\text{Ba}_2\text{Cu}_3\text{O}_4\text{Cl}_2$ but must be oxide origin $\text{Ba}_2\text{Cu}_3\text{O}_{6-x}$. Figure 7 shows temperature dependence of resistivity in the case of (b) measured by 4 probes in a cryogenic refrigerator. As temperature decreased, the resistivity increased showing semiconductor-like behaviour. At 45.5 K, superconductive transition was clearly observed. The samples (a) did not show superconductive transition.

4. Discussion

The diffraction patterns shown in figures 6(b) and (c) tell existence of the mixture of c-Y123 and a-Y123. In the reference [5] which is study of Y123 deposition by FF-MOD on STO (100) substrate, the similar combination of the X-ray diffraction pattern in figure 5 (c) and the temperature dependence of resistivity in figure 7 was shown. This fact indicates a metallic temperature dependence of resistivity and superconducting transition around 90 K caused by pure c-Y123 may be obtained with decreasing O_2 partial pressure. As for another possibility, we may get c-Y123 for more enhanced crystallization at higher annealing temperature. However, with increase in the annealing temperature, x-ray diffraction peaks of BaCeO_3 appeared as shown in figure 6(b) and (c), indicating that annealing temperature should be reduced. Although further experiments are indispensable, but a possibility of the fact that the sputter-deposited CeO_2/YSZ buffer layer on Si (100) substrate with

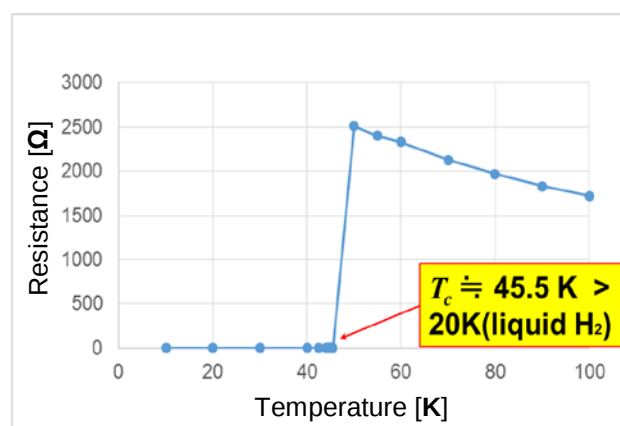


Figure 7. Temperature dependence of resistivity of the sample (c).

additional thin sputter-deposited Y123 film might have played the similar role on c-Y123 layer formation at relatively lower temperature might be brought about for further inspection.

5. Conclusion

By using the propionate FF-MOD solution with addition of DCA, a mixture of c-Y123 and a-Y123 film which shows superconducting transition at 45.5 K was obtained on CeO₂/YSZ buffered Si (100) wafer. These results were in good agreement with previously reported ones for FF-MOD on STO (100) substrate. From this previous literature, further reduction of O₂ partial pressure may lead to pure c-Y123 film formation. This results also teaches the propionate FF-MOD solution which was successful to form c-Y123 film on the steel substrate also works on CeO₂/YSZ buffered Si (100) substrate. By further adjustment of the experimental condition, we expect to deposit c-Y123 film on the Si wafers with spiral trenches as shown in figure 2 for a significant improvement of our compact SMES.

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