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Nb_3Sn THIN FILM DEPOSITION ON COPPER BY DC MAGNETRON SPUTTERING*

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Abstract

Nb_3Sn for SRF cavities has been coated on copper samples by DC magnetron Sputtering. Pure Nb and Sn target were installed separately in the magnetron sputtering device. Nb_3Sn precursor was coated on copper in the Ar atmosphere of 0.5 Pa. The Nb_3Sn precursor was annealed in the vacuum furnace whose pressure is 10^{-4} Pa. The XRD results demonstrate the exist of Nb_3Sn crystal, and MPMS results show superconductivity of Nb_3Sn. The highest critical temperature obtained is 15 K.

INTRODUCTION

Nb_3Sn is a promising alternative SRF material to bulk niobium. Its highest critical temperature reported is 18.3 K [1] and superheating field is 400 mT [2]. These two features are beneficial to SRF accelerator, including the high Q_0 of cavities, higher theoretical maximal accelerating field, comparing with bulk niobium cavity.

Copper cavities has several advantages, including better thermal stability (resistance to "quench") owing to the much higher thermal conductivity of OFE copper substrate and reduced material cost [3]. All of these advantage is compared with bulk niobium cavity.

Some researchers have demonstrated the feasibility of coating Nb_3Sn by magnetron sputtering. Rossi [4] deposited multiplayer Nb_3Sn on niobium samples and cavities by UHV magnetron sputtering technique. Rosaz [5] condensed Nb_3Sn film on copper sample by using a stoichiometric Nb-Sn target. Guitron [6] deposited Nb_3Sn films on sapphire, whose SRF properties has been characterized by SIC system.

To take advantage of the merits of Nb_3Sn and copper, Nb_3Sn thin film deposition on copper by dc magnetron sputtering is conducted in Peking University. Separated Nb and Sn target is used in magnetron sputtering device.

EXPERIMENT DETAILS

The Nb_3Sn films on copper coating is carried out on a DC magnetron sputtering device, which is shown in Fig. 1. Nb target and Sn target were installed in the vacuum chamber. The RRR of Niobium is above 300. The purity of Sn target is 99.99%. The sample holder is placed between two target. The chamber base pressure is 5×10^{-4} Pa. As the sample holder rotate, the Nb particle and Sn particle condense on the OFE copper substrate.

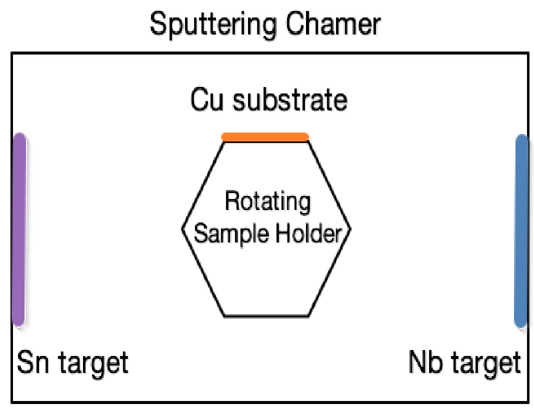

Figure 1: schematic picture of sputtering chamber. The Sn and Nb target were placed separately and Cu substrate is rotating.

The size of OFE Copper substrate is 25 mm ×50 mm. After mechanical polishing with 300# 600#, 1200#, 2000# sandpaper, chemical polishing is used. The polishing agent is SUBU solution [7] and working temperature is 72 °C. The SUBU is preceded for 30 minutes and followed by 10 minutes passivation with a dilute solution of sulfamic acid [8]. After mechanical and chemical polishing, the copper substrate is cleaned in ultrapure water by ultrasonic cleaning. The polished copper substrate is shown in Fig.2.

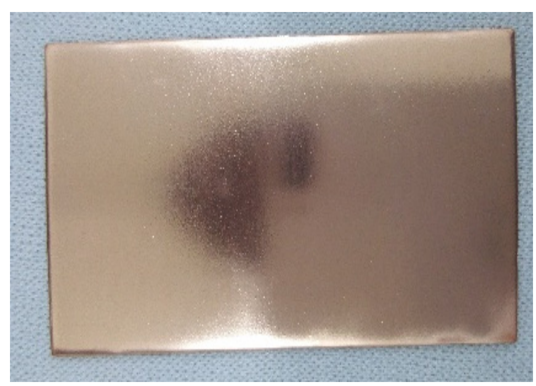

Figure 2: the copper substrate polished by SUBU solution.

The parameters of coating procedure are listed in Table 1. Isolating layer over the copper in used to avoid the reaction between Sn and Cu. In order to reduce contamination of other elements, the element Nb is chosen for isolating layer. After 6 min coating of isolating layer, the power of tin target is turn on and the sample begin to rotate. About 1 um Nb_3Sn precursor film on copper is obtained after the 240 min coating of mixing layer including Nb and Sn element. Sputtering process gas is Ar and the pressure maintains 0.5 Pa. The bias voltage added in copper substrate maintains -100 V.

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Table 1: Coating Parameters in DC Magnetron Sputtering

Sputtering Layer:	**Isolating Layer**	**Mixing Layer**	
Element:	Nb	Nb	Sn
Current (A):	1.20	1.20	0.15
Voltage (V):	295	295	325
Time (min):	6	240	
Bias Voltage (V):	-100	-100	

Nb_3Sn precursor is placed in quartz-tube vacuum furnace whose base pressure is $1\times10^{-4}\ Pa$. The annealing curves is given is Fig. 3. The temperature are initially ramped up to a stage of 200 °C for 2 hours in order to degas the furnace. Then the temperature is ramped up to the stage of 640 °C which lead to the formation of Nb_3Sn crystal. After annealing, the furnace is cooled naturally. The ramp rate of the whole heat treatment schedule is 20 °C/min.

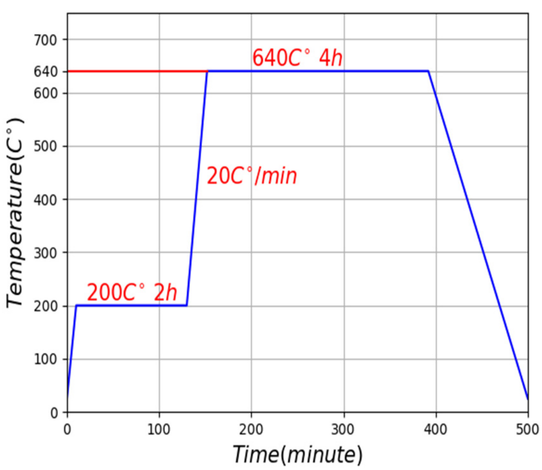

Figure 3: Temperature-time curves of annealing process. The stage of 200 °C for 2 hours is meant to degas the furnace.

After annealing, the crystal structure of Nb_3Sn is evaluated by using XRD method. The DC superconductivity of Nb_3Sn is measured by using Magnetic Property Measure System (MPMS). The surface morphology of Nb_3Sn film is observed by SEM and element ratio of Nb_3Sn is measured by EDX.

RESULTS AND DISCUSION

The annealing process is critical to the quality of Nb_3Sn. In order to analyze the influence of annealing temperature and time on the formation of Nb_3Sn. Experiments with different annealing temperature and time is carried out and those Nb_3Sn samples is evaluated by XRD and MPMS. Figure 4 shows the XRD diffraction patterns.

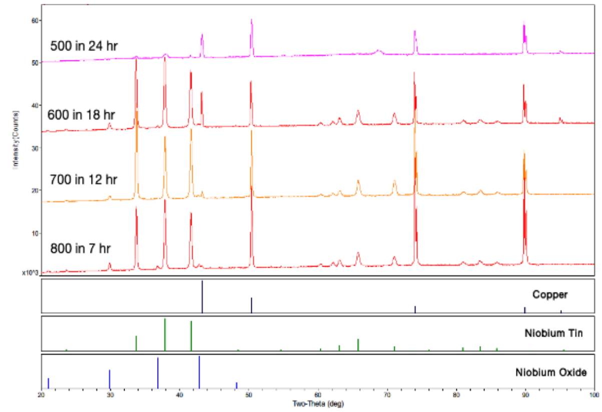

Figure 4: The XRD diffraction patterns of different annealing condition in the formation of Nb_3Sn.

From Fig. 4, if the annealing temperature is below 500 °C, the Nb_3Sn is amorphous which can't become superconducting. When the temperature raise to 600 °C or more, Nb_3Sn polycrystalline exists. No distinct differences can be observed among those XRD results which exist Nb_3Sn polycrystalline even the annealing temperature and time is quite different. Besides, there exist Niobium Oxide which may related to the Nb isolating layer or excessive Nb in Nb_3Sn film.

DC superconductivity of Nb_3Sn samples were measured by MPMS. The principle of MPMS is that measuring the magnetic moment of samples. When the superconducting samples quenched as the temperature increase, its negative magnetic moment become nearly zero. By placing the superconducting sample in a small constant magnetic field, the sample is diamagnetism. Through measuring the magnetic moment of samples in different low temperature, the dc superconductivity of samples is obtained. The influence of annealing time and temperature on DC superconductivity of Nb_3Sn samples is given in Fig. 5. The maximum critical temperature become bigger as the annealing time and temperature increase. The MPMS results demonstrate there exist superconductors with different critical temperature from 2 K to 15 K.

One explanation of such phenomenon is that the critical temperature of Nb_3Sn is related to the ratio of Sn [9]. As shown in Fig. 6. The ratio Sn in Nb_3Sn could be 18~25% and corresponding Tc is 6 K-18 K. This means excessive Nb or less Sn will induce the decrease of Tc.

The SEM picture of Nb_3Sn is given in Fig. 7. This Nb_3Sn samples were annealing in the 640 °C 20 h. The SEM pictures show some cracks in the film which indicate the fragility of Nb_3Sn.

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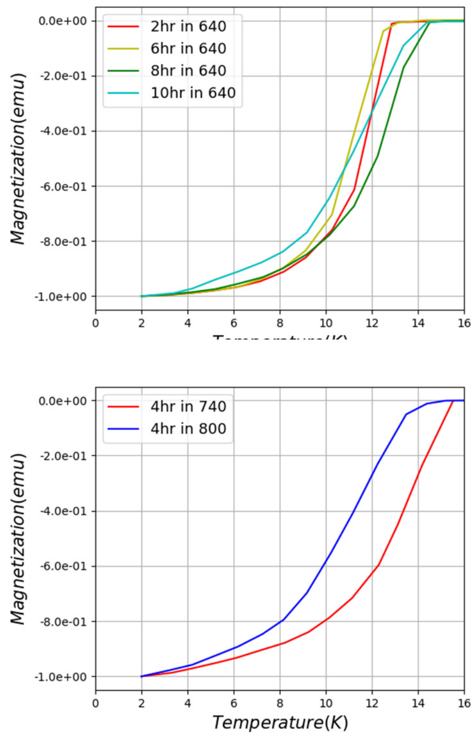

Figure 5: The MPMS results of Nb_3Sn sample annealing different time and temperature.

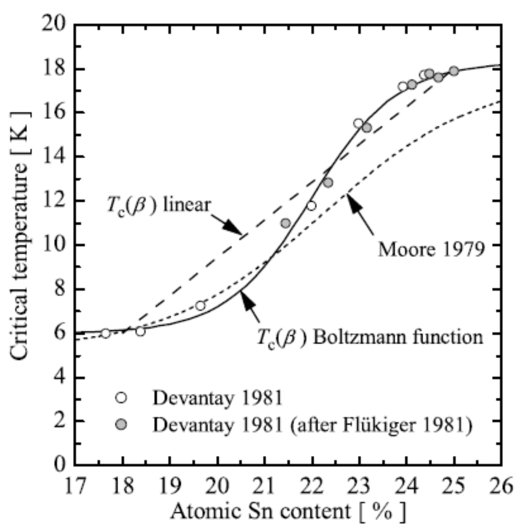

Figure 6: Literature results for the critical temperature as functions of Nb-Sn composition.

EDS was used to evaluate the element ratio in Nb_3Sn film. In EDS technique, electron beam of specific energy is used to induce the characteristic X-ray of element in the film. Table 2 shows the results of EDS. From Fig. 7, it's obvious that the ratio between Nb and Sn is bigger than 3, this results is consistent with XRD results and MPMS results.

A significant problem in annealing indicated by EDS results is that the distribution of Sn in film is change along the depth. As the Table 2 shows, the amount of Sn is decreasing as the energy of electron beam decrease. The energy of electron beam decides the detect region along the film depth. During annealing, the amount of Sn near the surface evaporate much more than that deeper. Another problem is that the oxygen takes up most part of all element, which may influence the rf performance of thin film.

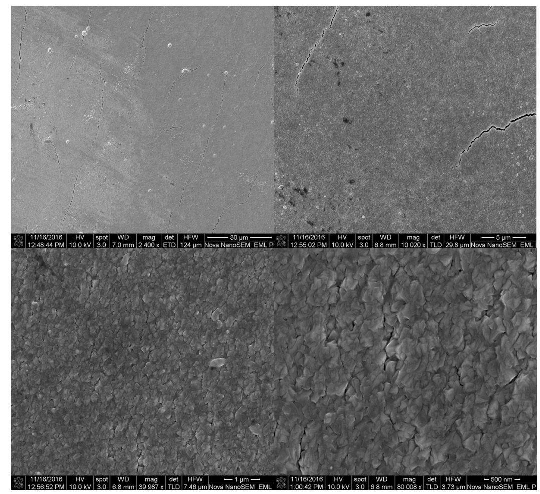

Figure 7: SEM picture of Nb_3Sn films annealing 640℃ 20h. It's obvious to see some cracks on the film.

Table 2: EDS Results of Nb_3Sn Film. The Ratio Between Nb and Sn is Bigger Than 3. The Amount of Sn is Decreasing as The Energy of Electron Beam Decrease. The Amount of Oxygen is Biggest.

Energy	**20 keV**		**10 keV**			**5 keV**
Nb Atom %	32.10	32.33	34.70	34.17	34.03	35.78
Sn Atom %	6.40	6.34	2.89	2.97	2.98	0.00
O Atom %	53.26	52.70	55.94	56.21	56.43	63.14
Cu Atom %	8.24	8.64	6.47	6.65	6.56	1.08

CONCLUSIONS

Nb_3Sn for SRF cavities has been coated on copper samples by DC magnetron Sputtering. This experiments demonstrate the feasibility of coating Nb_3Sn on copper by sputtering with two separate Nb and Sn target. The XRD diffraction patterns indicated the exist of Nb_3Sn crystal. The superconductivity of Nb_3Sn film on copper has been proved by MPMS results. The highest critical temperature obtained is 15K.

Some problems need to be alleviated including excessive Nb and O, cracks of Nb_3Sn film, and the non-uniform distribution of Sn along the film depth.

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REFERENCES

[1] J.J. Hanak, K. Strater, G.W. Cullen, "Preparation and properties of vapor-deposited niobium stannide", *RCA (Radio Corporation of America) Review (US).*, vol. 25, 1964.

[2] J. Matricon, "Saint-James D. Superheating fields in superconductors", *Physics Letters A.*, vol. 25, no. 5, pp. 241-242, 1967.

[3] S. Calatroni, "20 Years of experience with the Nb/Cu technology for superconducting cavities and perspectives for future developments", *Physica C: Superconductivity.*, vol. 441, no. 1–2, pp. 95–101, Jul. 2006.

[4] A.A. Rossi *et al.*, "Nb_3Sn films by multilayer sputtering", *Proceedings of SRF2009,* Berlin, Germany, paper TUOBAU06, pp. 149-154.

[5] G. Rosaz *et al.*, "Development of Nb_3Sn coatings by magnetron sputtering for SRF cavities" *Proceedings of SRF 2015*, Whistler, BC, Canada, paper TUPB051, pp.691-694.

[6] S. Sosa-Guitron, *et al.*, "Measurements of RF properties of thin film Nb3Sn superconducting multilayers using a calorimetric technique", *Proceedings of SRF 2015*, Whistler, BC, Canada, paper TUPB060, pp. 720—722.

[7] J.-P. Birabeau, J.M.A. Guerin, "BAIN DE POLISSAGE CHIMIQUE DE METAUX ET ALLIAGES DE METAUS", Patent No 8809820, Institut National de la Proprie´te´ Industrielle, 1993.

[8] C. Benvenuti *et al.*, "Study of the surface resistance of superconducting niobium films at 1.5 GHz", *Physica C: Superconductivity.*, vol. 316, no. 3, pp. 153-188, 1999.

[9] A. Godeke, "A review of the properties of Nb3Sn and their variation with A15 composition, morphology and strain state", *Superconductor Science and Technology*, vol. 19, no. 8, 2006.