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SEVERAL EXPERIMENTAL PHENOMENA OF Sn NUCLEATION ON Nb SURFACE OBSERVED AT IMP*

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Abstract

Nucleation process is an important step that affects the quality of Nb_3Sn films coated by vapour diffusion method. A uniform distribution of nucleation centres is essential to the uniformity of Nb_3Sn films. In this paper we report several experimental phenomena on the Sn nucleation on Nb surface. Better nucleation in the downstream of the pumping direction was observed. Influence of $SnCl_2$ partial pressure inhomogeneity was studied. Samples with higher $SnCl_2$ partial pressure have denser nucleation, which means homogeneous $SnCl_2$ pressure is a critical factor to the uniform nucleation. Less-nuclear zones, mainly distributed at cracks, grain boundaries and even some whole grain surfaces, were found on the surfaces of all samples. The less-nuclear zones may result in the low tin regions of the Nb_3Sn cavities. The specific solution to the less-nuclear problem was proposed. These studies help to better understanding of the mechanism underlying the nucleation process and will be useful foundation for the follow-up Nb_3Sn/Nb project at IMP.

INTRODUCTION

After many years of worldwide exploration and improvement of the post treatment process, niobium cavities are now approaching their fundamental limits both in terms of maximum accelerating field as well as in terms of surface resistance at typical operating temperatures. To continue to keep up with continually increasing demands of future SRF facilities, there exists urgent demand for the development of new technologies for RF superconductivity.

The A15 superconductor Nb_3Sn is one of the most promising alternative materials to standard niobium for SRF applications not only for its material parameters with obvious advantages [1] but also for the breakthrough performance levels achieved for the vapour diffusion coated 1.3 GHz single cell cavity at Cornell University [2].

One key factor limiting the RF performance of the vapor diffusion coated Nb_3Sn thin film cavities is the heterogeneity of film composition and distribution. An important manifestation of inhomogeneity is the existence of areas with low tin content [2-3]. Presently, the main efforts to further improve the RF performance of the vapour diffusion coated Nb_3Sn thin film cavities is through the optimization of the coating parameters at high temperature, however [2, 4], its effects is not obvious. In the past three years, the RF performance of the Nb_3Sn thin film cavities has not been substantially improved [2, 3, 5].

Whether it is from the growth mechanism of vapour diffusion coated Nb3Sn film, or from the research experience of Siemens AG researchers [6], a uniform distribution of nucleation centres is essential to the uniformity of Nb_3Sn films. Without uniform nucleation, it is difficult to avoid the heterogeneity of Nb_3Sn film composition and distribution even with the optimization of the coating parameters at high temperature. Compared to the optimization of the coating parameters at high temperature, the importance of the nucleation process to break through the limitations of the RF performance of Nb_3Sn cavities has not been paid sufficient attention, and research on nucleation [7] is relatively less.

Under such a background, a thorough study of Sn nucleation on Nb surface was carried out at IMP, which will be useful foundation for the follow-up Nb_3Sn/Nb project at IMP. In this paper, several experimental phenomenon of Sn nucleation on Nb surface will be reported.

EXPERIMENTAL

Sample Preparation

The Nb samples were cut by wire electro-discharge machining (EDM) from the same plate. The samples' treatments were attempted to replicate that of the cavities, which includes polishing smoothly on 1000-grit sandpaper, 150μm heavy BCP (HF: HNO_3: H_3PO_4 =1:1:2), 8000C degas for 3 hours and 20μm light BCP.

Nucleation Experiment

The nucleation experiment of the niobium samples was carried out in a furnace using a high purity quartz tube, taking the advantage of high purity quartz's good thermal and chemical stability at high temperature. The furnace's background pressure is better than $5\times10^{-}5$ Pa, which can meet the requirement of nucleation experiment. The nucleation was accomplished by using the nucleation agent $SnCl_2$. After 24 hours of degasing at 200^0C in high vacuum, the temperature was raised up to 500^0C at a rate of 3.50C/min. Then the samples were kept at 500^0C for 4 hours for nucleation. After which, the furnace was turned off to cool down naturally.

RESULTS

Influence of Pumping Direction

The nucleation was accomplished taking the advantage of $SnCl_2$'s high vapour pressure at relatively low temperature. According to common sense, the $SnCl_2$ molecule moves downstream along the pumping direction. This will possibly cause that the partial pressure of $SnCl_2$ in the downstream of the pumping direction is larger than that of the upstream of the pumping direction, resulting in the difference in the nucleation distribution on the surfac-

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es of the Nb samples in the upstream and downstream direction. To verify this conjecture, an experiment was designed with the size of pipe quartz plug placed both in the upstream and downstream of the pumping direction. The detailed experimental setup was shown in Fig. 1.

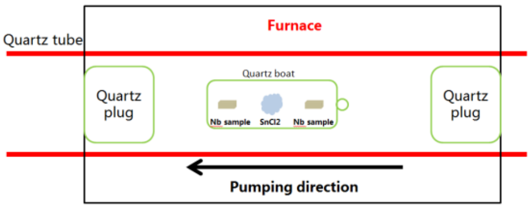
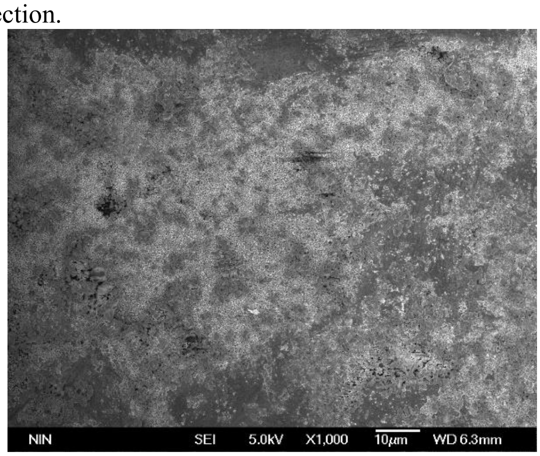
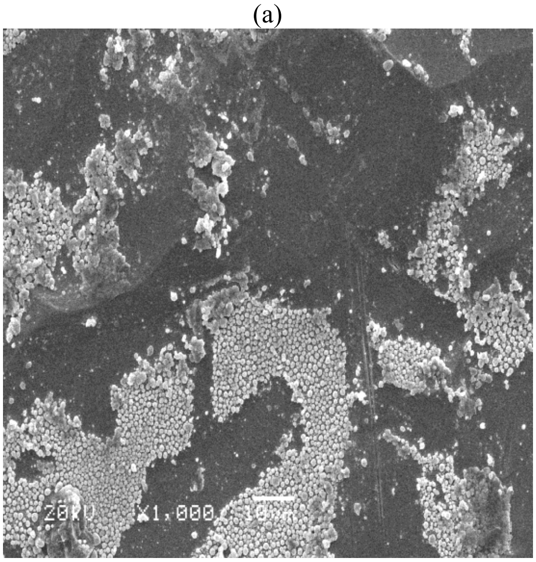


Figure 1: Detailed setup of the experiment to verify the effects of pumping direction.

The SEM images in Fig. 2 directly show that there is denser and better nucleation in the downstream pumping direction.



(a)



(b)

Figure 2: Nucleation of Nb samples in the downstream (a) and upstream pumping direction to study the influence of pumping direction.

Influence of Aeration Condition

To further investigate the effect of $SnCl_2$ partial pressure uniformity on nucleation, another series of experiments were designed with the detailed conditions shown in Fig. 3. A larger pipe quartz plug was placed in the downstream of the pumping direction. In this case, although the $SnCl_2$ molecule still has the desire to move downstream along the pumping direction, due to the blocking of the larger pipe quartz plug in the downstream, the difference of the partial pressure of $SnCl_2$ between the upstream and downstream of the pumping direction will be effectively reduced, and even the partial pressure of $SnCl_2$ in the upstream direction of the pumping direction may be larger.

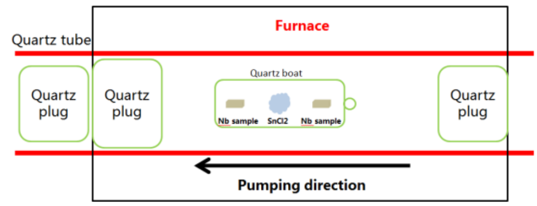
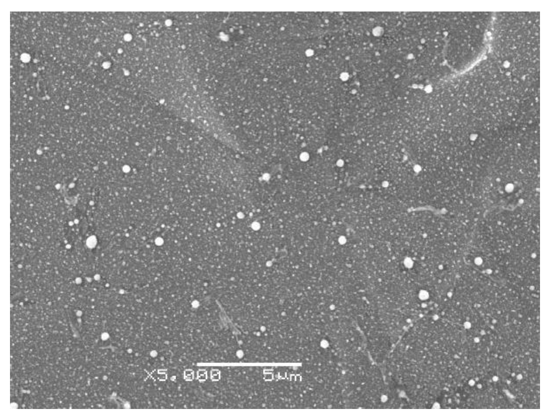
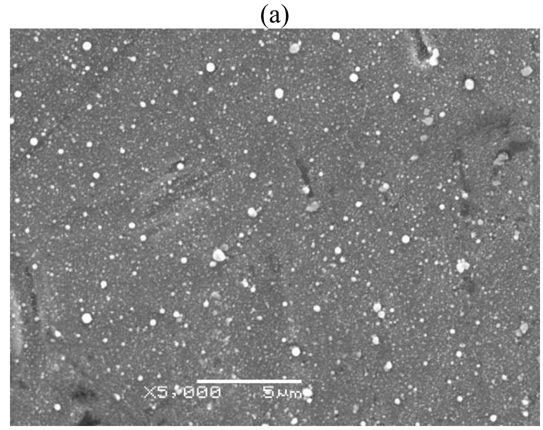


Figure 3: Detailed setup of the experiment to verify the effects of aeration condition.

SEM images of the nucleation Nb samples in the downstream and upstream pumping direction with the $SnCl_2$ dosage of 100mg were shown in Fig. 4 for reference. From the naked eye observation, different from the nucleation in "Influence of pumping direction" part, the Nb sample in the upstream pumping direction seems to have denser nucleation centres.



(a)



(b)

Figure 4: Nucleation of Nb samples in the downstream (a) and upstream (b) pumping direction to study the effect of aeration condition.

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EDS results of Nb samples both in the downstream and upstream pumping direction with various amounts of $SnCl_2$ dosage was listed in Table 1.

Table 1: EDS Results of Nucleation Nb Samples

Experiment	Sn At %-downstream	Sn At %-upstream
400mg $SnCl_2$+Sn	6.86	9.61
100mg $SnCl_2$	3.03	4.11
400mg $SnCl_2$+Sn	2.31	2.81
11.9mg $SnCl_2$	1.31	1.64
11.8mg $SnCl_2$+Sn	1.27	1.27

The EDS results showed that in the case of the larger pipe quartz plug placed in the downstream pumping direction, all the Nb samples in the upstream pumping direction have higher concentration of Sn on the surfaces. This means the samples in the upstream pumping direction have denser nucleation centres, which implies the higher partial $SnCl_2$ in the upstream pumping direction. This agrees with the tentative presumption.

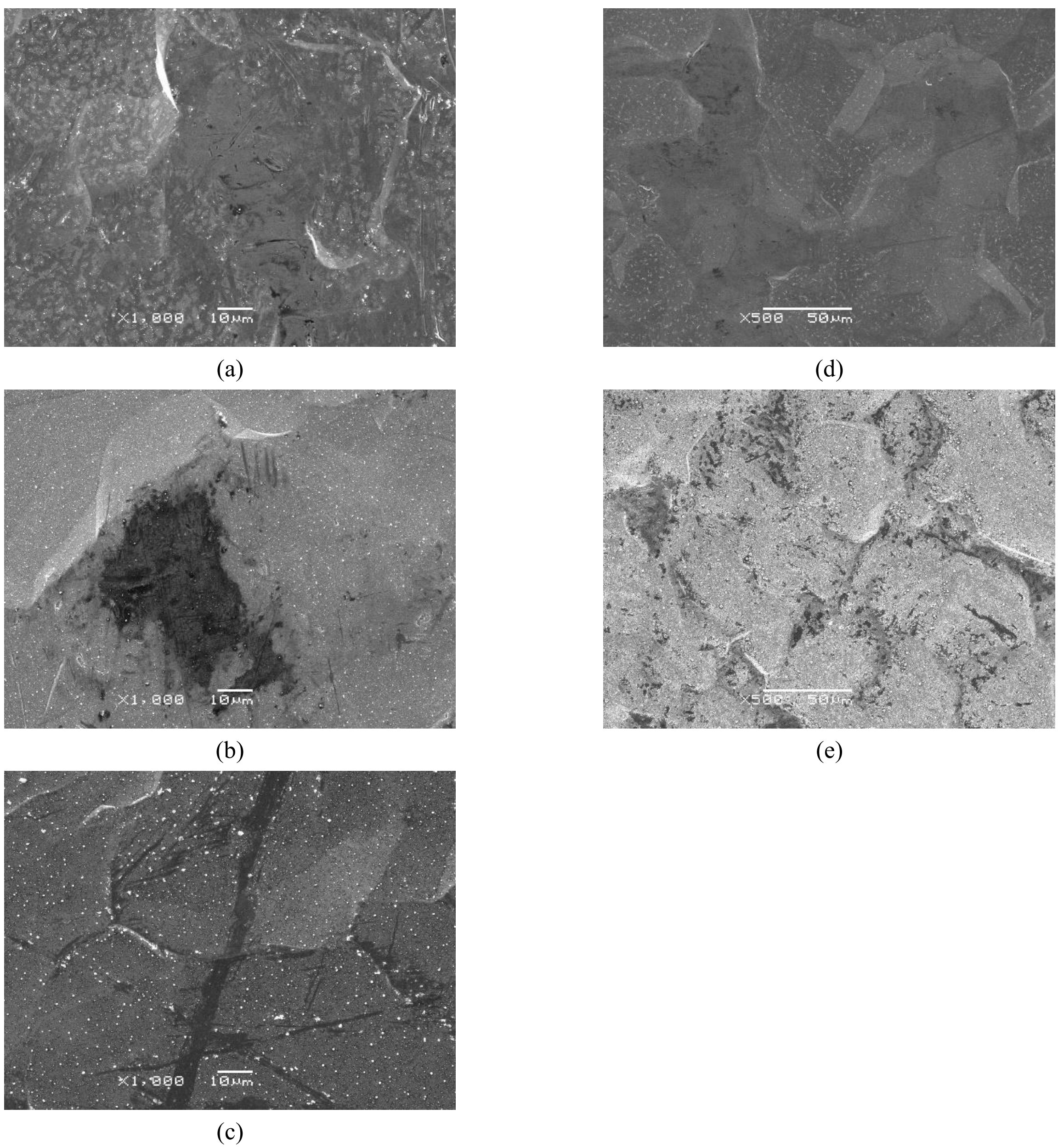

Figure 5: Less-nuclear zones in Nb nucleation samples with $SnCl_2$ dosage of 11.9mg (a) and 100mg (b). Less-nuclear zones at scratches (c), grain boundaries (e) and even some whole grain surfaces (d).

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Table 2: EDS Results of Both the Less-nuclear Zones and the Zones with Uniform Nucleation Centres

Experiment	Sn At % of left-uniform nucleaton	Sn At % of left-less-nuclear	Sn At % of right-uniform nucleaton	Sn At % of right-less-nuclear
400mg $SnCl_2$+Sn	6.86	4.94	9.61	2.10
100mg $SnCl_2$	3.03	1.04	4.11	1.34
400mg $SnCl_2$+Sn	2.31	1.25	2.81	1.21
11.9mg $SnCl_2$	1.31	0.86	1.64	1.16
11.8mg $SnCl_2$+Sn	1.27	1.52[1]	1.27	0[2]

[1, 2] Proximity of EDS limit of detection accuracy

Observation of Less-nuclear Zones

As stated in the introduction part, the existence of areas with low tin content is an important factor limiting the RF performance of the vapour diffusion coated Nb_3Sn thin film cavities. The reason and mechanism of the occurrence of the less-tin areas remain unclear, neither the way to restrain it.

During the study of Sn nucleation on Nb surface at IMP, unexpected areas of less nucleation centres were observed at the surfaces of all nucleation Nb samples. The specific distribution of the less-nuclear zones is shown in Fig. 5.

SEM images showed that the less-nuclear zones mainly distributed at scratches, grain boundaries and even some whole grain surfaces. It implies that a smoother surface is beneficial to a uniform nucleation and defects such as pits and scratches should be avoided as much as possible. In addition, the most important information reflected in the observed less-nuclear phenomenon is that the less-nuclear zones even cover some whole grain surfaces, which means that nucleation by $SnCl_2$ agent seems to have crystal direction selectivity.

In order to prove that the observed less-nuclear zones in the SEM images really has less nucleation centres, EDS was performed to measure the exact Sn content both in the less-nuclear zones and the zones with uniform nucleation centres. The results were listed in Table 2. Recently, the presence of less-nuclear zones has also been observed on the surface of the latest batch of nucleation Nb samples. The location of the less-nuclear zones is still the scratches, the grain boundaries and even some whole grain surfaces.

CONCLUSION

From the research status of the past few years, the importance of the nucleation process to break through the limitations of the RF performance of Nb_3Sn cavities has not been paid sufficient attention, and research on nucleation is relatively less. The specific structural design of the furnace for the coating of the Nb_3Sn thin film cavities by the vapour diffusion method also lacks corresponding specifications. The observed influence of pumping direction and aeration condition on the Sn nucleation on Nb surfaces by using the $SnCl_2$ agent clearly showed that a homogeneous SnCl2 pressure is a critical factor to the uniform nucleation. The observed influence of pumping direction and the aeration conditions provides normative guidance for the design and improvement of the furnace, the location of $SnCl_2$ agent and the optimization of nucleation conditions.

Less-nuclear zones were observed on all the nucleation Nb samples. The observed less-nuclear zones mainly distributed at the scratches, the grain boundaries and even some whole grain surfaces. The occurrence of less-nuclear zones at the scratches and the grain boundaries implies that a smoother surface is beneficial to a uniform nucleation. The defects on the inner surface of the substrate Nb cavity such as pits and mechanical scratches must be avoided as much as possible. The information revealed by the presence of less-zones at some whole grain surfaces is more important. It seems that nucleation by $SnCl_2$ agent have crystal direction selectivity. The occurrence of less-nuclear zones at some whole grain surfaces seems to be ineluctable. From the growth mechanism of vapour diffusion coated Nb_3Sn film, the occurrence of the less-nuclear zones will result in the locally too thin Nb_3Sn film or localized Sn deficiency, both of which lead to increased losses. What's more, the size of the less-nuclear zones at the whole grain surface is comparable to that of the less-tin zones reported by Cornell University. The less-nuclear zones may be an important source of the reported less-tin zones. However, in any case, suppressing the occurrence of less nuclear regions will reduce the heterogeneity of the vapour diffusion coated Nb_3Sn films and may be a possible way to break through the RF performance limitation of the Nb_3Sn cavities.

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