EFFECTS OF CATHODE SHAPES ON BEP AND EP DURING VERTICAL SURFACE TREATMENTS ON NIOBIUM*

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Abstract

This paper reports the research results of effects of cathode shapes during buffered and conventional vertical electropolishing treatments for single cell superconducting radio frequency (SRF) niobium cavities. Several different cathode shapes such as, for instance, bar, ball, ellipsoid, wheel, etc. were employed. Detailed electropolishing parameters at different locations inside a single cell SRF cavity were measured using a unique JLab home-made demountable cavity, including I-V characteristic, removal rate, surface roughness, polishing uniformity and so on. It was demonstrated that optimal polishing results could be achieved by changing the cathode shape for both BEP and EP. Implications on the electropolishing mechanism of Nb cavities for both BEP and EP based on the obtained experimental results are discussed.

INSTRUCTION

Previous study shows that reactions at different locations of the cavity may be different especially during BEP process [1]. This paper mainly focused on the effect of cathode shape on the process of BEP. Detailed parameters at different locations inside the demountable cavity such as, I-V characteristic, removed rate, surface roughness, polishing uniformity, and so on were measured by a demountable cavity. It was revealed that cathode shape had dominant effects on the inhomogeneous polishing rates between the equator and iris in an Nb SRF single cell cavity for buffered electropolishing (BEP). The conventional electropolishing (EP) appeared to have the same tendency. This study demonstrated that a more homogeneous polishing result could be obtained by optimizing the electric field distribution inside the cavity through the modification of the cathode shape given the conditions that temperature and electrolyte flow were kept constant.

EXPERIMENTAL SETUP

The experimental setup mainly includes four parts: 1. Demountable cavity; 2. Electrolyte circulating system; 3. Data acquisition system; 4. Cooling system. With the chemical fume hood, they constitute the whole setup. Besides, several cathodes of different shapes are tried as now in figure 1. More details please see the paper TUPO033 in this proceeding.

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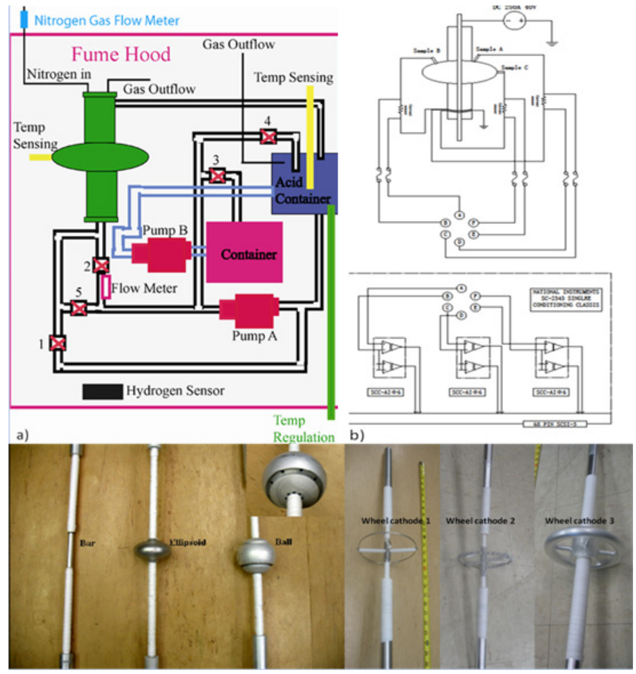

Fig. 1: Schematics of (a) the flow chart of BEP system and (b) the data acquisition system of the vertical BEP system for Nb SRF single cell cavities and the cathodes used in this study.

RESULTS AND DISCUSSION

Effect of Cathode Shape on I-V Characteristic

An electropolishing system consists of mainly three parts: anode, cathode and electrolyte. So, research on the effect of cathode becomes one of critical points in the whole electropolishing treatment study. In this part, we will mainly discuss the effect of different cathode shape on the I-V characteristic, and try to find the way on how to apply the experimental findings in real EP and EP processes. The effect of cathode in BEP process will be firstly discussed, and then we will talk about EP process.

This study was done through the measurements of a series of I-V curves with different cathode shapes in BEP experiments. In the experiments of BEP-2, BEP-6, BEP-8, BEP-13 and BEP-14, the cathodes used were thin bar, ellipsoid, ball, wheel cathode 3 and thick bar cathode, respectively. Most of them are shown in figure 1. As shown in figure 2, I-V curves from the different cathodes show great differences. They can be obviously distinguished in the etching region. The one with the smallest slope in the etching region is obtained by thin bar cathode, and then is obtained by thick bar cathode, ellipsoid cathode, ball cathode. The I-V curve which has the largest slope in the etching region was obtained by wheel cathode 3. Then with the increase of voltage, we can find that the I-V curves obtained by the thin bar, thick bar and ellipsoid cathode in experiments BEP-2, BEP-14, BEP- 6, respectively, don't have an obvious best polishing region. For the ball shape cathode, although its I-V curve has the polishing region, its oscillation region is a smooth transition. Only the I-V curve obtained by wheel cathode 3 has the whole typical regions. We think those differences should come from following two different factors: initial electric field distribution and cathode surface area which was shown in the table 1.

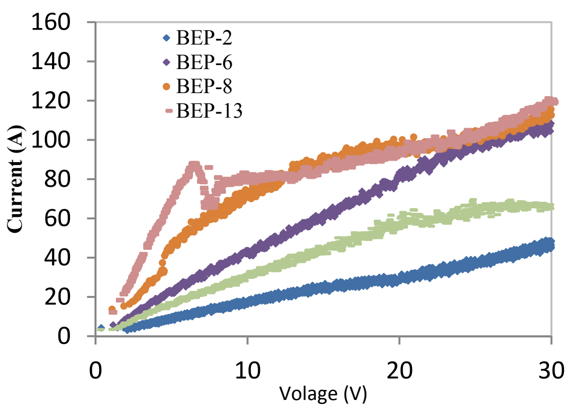

Figure 2: The cavity I-V curves from the different cathode shapes in BEP experiment series 1 (BEP-2: thin bar cathode; BEP-6: ellipsoid cathode; BEP-8: ball cathode; BEP-13: wheel cathode 3; BEP-14: thick bar cathode).

First, for the results of experiment BEP-2, BEP-6 and BEP-14 discussed above, we think the main reason was due to the differences of cathode areas. As we know, in electropolishing process, the cathode will produce a large amount of hydrogen gas which will form a gas curtain around it in the electrolyte. Since the quantity of H_2 is determined by the current in the reaction, with the same current the cathode with a smaller surface will have a thicker layer which we call it "gas curtain around the cathode", and then the more voltage will be dropped by the gas curtain. So, if we want to get the same potential drop on the anode, the whole voltage between anode and cathode will be larger for the smaller cathode surface. Apart from creating the drop, the gas curtain also has the effect to prevent the ions from getting to the cathode surface. So, the cathode polarization also moves the I-V towards the higher voltage area.

As to prove this assumption about the cathode area effect, the cathode area research was carried out with small sample experiments. The result is shown in figure 3, in which the percentage represents the ratio of cathode area to anode area. As we see, the same trend was obtained in the small flat sample experiments as that shown in figure 3. When the cathode area is below a

Table 1: Ratios of initial electric field between sample 3 and sample 1 and electrode surface area between cathode and anode for different cathode shapes.

Experiments	Cathode	Electric field ratio between sample 3 and 1	Area ratio between cathode and anode
BEP-2	thin bar	0.13	2.96%
BEP-6	ellipsoid	0.21	12.68%
BEP-8	ball	0.16	16.90%
BEP-13	wheel 3	1.06	54.23%
BEP-14	thick bar	0.13	7.75%

specific ratio about 7% in BEP process with respect to anode, the I-V curves even cannot have the polishing plateau below 30V. However, the minimum area of cathode required in the cavity experiments shows a little larger than that in the sample experiment. This is not hard to understand since the reaction environment in the cavity experiment is closed, and the electrolyte volume is also less comparing to that in the sample experiment. So, the effect of hydrogen gas will be greater in the cavity polishing process than that in the sample experiment. The same experiment about surface area research was also carried out with EP process. We found the tendency of the I-V curves' development in EP process was similar to that in BEP process. However, there was still a little difference in EP since we observed that there was a more obvious oscillation phenomenon even in the plateau region. We thought that this might be caused by the different specific mechanisms of reaction between BEP and EP due to the difference of electrolytes.

For the difference between the I-V curve obtained by the ball cathode (shown as BEP-13 in figure 2) in BEP experiment and the typical electropolishing I-V curve, we think that the problem mainly comes from the inhomogeneous initial electric field distribution. Here, the

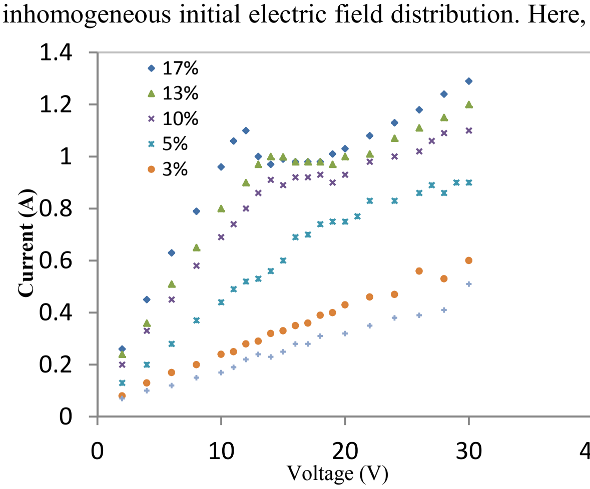

Figure 3: The I-V curves obtained from small sample experiments with different cathode area during BEP. The percentages shown in the figure are the surface area ratios between cathode and anode.

initial electric field distribution is the electric field

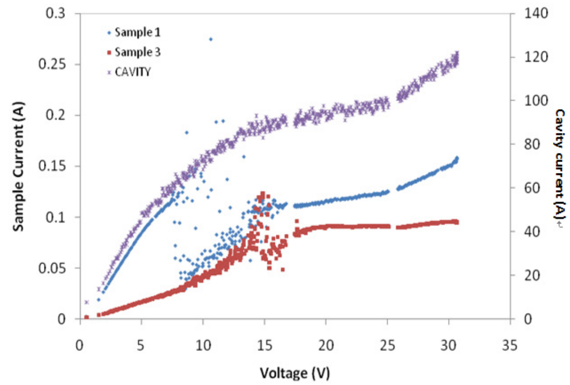

Figure 4: The I-V curves in the BEP experiment using the ball shape cathode.

distribution in etching region before the compact solid layer formed or diffusion effect start to play a role [2]. Due to the initial non-uniform electric field, the layer also cannot form at the same applied voltage for all the inner surface of the cavity at the same time. In figure 4, we can see this phenomenon. The compact solid layer of sample 1 is formed at 9V, while the voltage is 14V for sample 3. So, the inconformity of the oxide layer forming voltages at different locations of the inner surface of a cavity causes a disappearance of the sharp maximum current point in the whole cavity's I-V curve. This phenomenon was also proved in the experiment with a wheel cathode 3. Figure 5 was the simulated initial electric field distribution with the wheel cathode 3. Since inner bar part for the wheel cathode 3 has four holders between the outer ring part, we have to do following two situations: with the holders and without the holders. The simulation result using Poisson-Superfish showed that it had a uniform electric field distribution for both situations (see Fig.6). I-V curves of cavity and two samples obtained by using the wheel cathode are shown in figure 6. One can see that with the more uniform initial electric field distribution, the oxide layer forming voltages of sample 1 and sample 3 are almost the same. So, maximum peaks of the button samples are situated at the same voltage as that of the cavity.

Figure 7 is a series of I-V curves obtained in EP

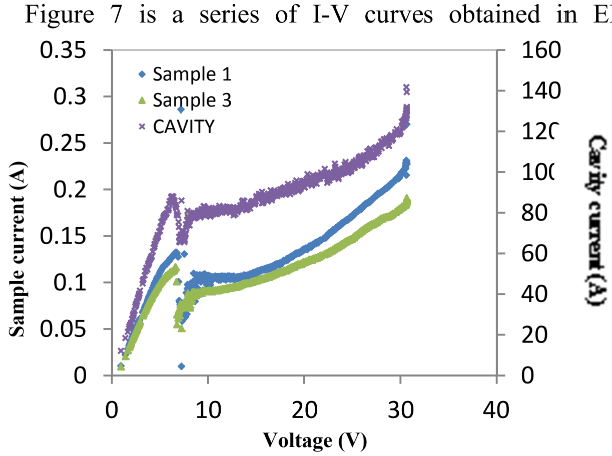

Figure 6: The I-V curves in BEP experiment using the wheel cathode 3.

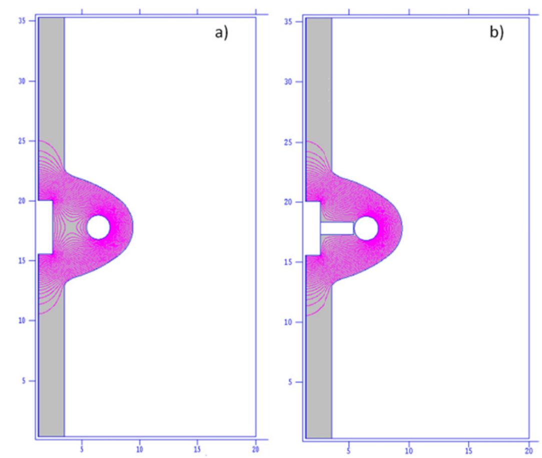

Figure 5: The simulated electric field distributions in the cavity with wheel cathode 3.

process with different cathode shapes. In the experiment EP-9, EP-10, EP-11, EP-12, cathodes of ball shape, thin bar shape, thick bar ship and wheel cathode 3 were used respectively. If we still distinguish them with the slope in etching region in the same fashion as those discussed for BEP, the I-V curve with the smallest slope was still obtained by the thin bar shape cathode, and then the larger slope for thick bar cathode, ball shape cathode, and the largest slope was obtained by using wheel cathode 3.

Comparing with BEP I-V curves, the developing tendency of those I-V curves in EP experiments is the same as that in BEP experiments. So, we think that this phenomenon is also due to the differences in cathode surface areas and the effect of gas curtain around the cathodes. However, we also can see some difference sbetween BEP and EP process from figures 3 and 8. One is that, in EP, the typical I-V curve is obviously easier to be obtained than that in BEP processes, especially for the appearance of maximum current peak in I-V curve. The other is that the voltage for the appearance of I-V curve maximum peak is lower in EP process than that in BEP process. For the first phenomenon, we think that it is due to the fact that the differences in the voltages for forming

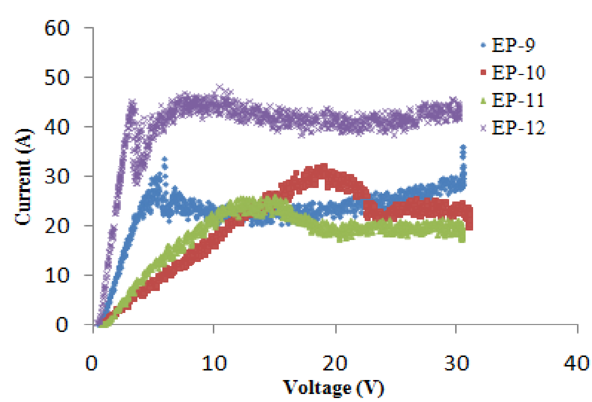

Figure 7: The Cavity I-V curves from the different cathode shapes in EP experiments (EP-9: ball cathode; EP-10: thin bar cathode; EP-11: thick bar cathode; EP-12: wheel cathode 3).

the compact solid layer in BEP processes are larger than their counterparts in EP processes as discussed above. The smaller differences of voltages for the forming of the oxide layer between different locations in EP process make the appearance of a maximum current peak in the cavity I-V curve easier. As to the phenomenon that the voltage for the current peak appearance is lower in EP process than in BEP, we think that, apart from the difference in electrolytes, the current in EP process is much smaller than that of BEP process as shown in figures 3 and 8. So, the gas generated in the polishing process will be less in EP than that in BEP process leading to the relative less voltage drop caused by bubbles in EP than that in BEP

Another thing we want to discuss here is the limited currents with the different cathode shapes in EP process. We can see, with the thin bar, thick bar and ball cathode, the limited currents in EP process are similar, which are around 20A to 25A. However, with the wheel cathode 3, the limited current is about 40A. We think it is because the electric field generated by wheel cathode 3 is more uniform and larger on the surface of the anode than that with the other cathodes. So, the larger electric field will increase the removal rate. This will be discussed in detail in the next section. The other explanation about this phenomenon is that the hydrogen bubbles generated by the wheel cathode will increase the flow rate in comparison with the other cathode shapes. However, we feel that the first reason is more likely since the hydrogen bubbles concentration is always higher for the area close to the iris than the equator; however, it does not show a faster removal rate in the button sample close to the iris with the wheel cathode as shown in table 2.

Effect of Cathode Shape on Removal Rate

Table 2: The ratios of removal rates between equator and iris from different cathode shapes in BEP and EP experiments

Cathod e	electric field ratio sample3/s ample1	removal rate ratio in BEP process sample3/sample1	removal rate ratio in EP process sample3/sampl e1
thin bar cathode	0.13	0.12 μm /0.57 μm =0.21	0.43 μm /0.85 μm =0.51
ball cathode	0.16	2.07 μm /4.28 μm =0.48	0.24 μm /0.48 μm =0.50
wheel cathode 3	1.06	3.15 μm /2.44 μm =1.29	0.72 μm /0.65 μm =1.16
thick bar cathode	0.14	1.84 μm /2.81 μm =0.65	0.17 μm /0.23 μm =0.74

Removal rate is one of most important parameters in the polishing process. It includes two aspects which need the most attention. One is about the absolute removal rate. The other is about the uniformity of the removal rate in a cavity. The removal rate has the greatest difference between iris and equator according to the previous experiences in horizontal EP when a simple bar shape cathode is used.

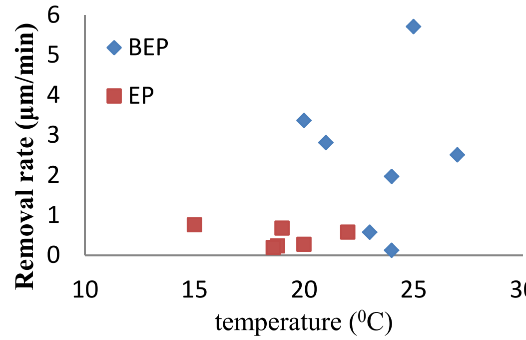

Figure 8: The relationship between temperature and removal rate in EP and BEP processes.

For the first aspect, the research on vertical polishing and BEP technology can lead to an increase in absolute removal rate. Especially for BEP, the research on small sample experiment had shown that it had much faster removal rate than that of EP. In this study, this result was confirmed again. Figure 8 shows the relationship between temperature and removal rate in BEP and EP processes corresponding to the experiments shown in table 2. For the two lowest points of the removal rate data in the BEP processes shown in the figure 8, both were obtained from the thin bar cathode, and we cannot get the right I-V curve since most voltage dropped in the hydrogen curtain around the small cathode surface area. So, those two points cannot reflect the real results. In this series of study, BEP still shows a much higher removal rate than that of EP. Besides, as shown in figure 8 from this serious study, in the range of 15^{o}C to 22 ^{o}C for EP and 20 ^{o}C to 27 ^{o}C for BEP, there are not an obvious relationship between the removal rate and temperature. This implies that temperature variation in this range does not show too much significant effect on the removal rate as compared with the effect from the changes in cathode shape.

Now, let us discuss about the other aspect of removal rate: polishing uniformity. For this topic, we will mainly use the ratio of the removal rates between sample 3 and sample 1 to try to understand reaction mechanism which causes the differences in removal rates between iris and equator. With the help from previous experience in horizontal EP, the removal rate at equator usually was half of the removal rate at iris. Similar phenomena were also found in our vertical electropolishing process including BEP and EP with most cathode shapes like bar, ball as well and thick bar cathode as shown in table 2. We think that the main reason for the different removal rate between sample 1 and sample 3 is from the different electric fields at the two different locations. As shown in

Figure 9: The CCD pictures of single cell cavity treated by EP process with the wheel cathode 3. Left one is the upper half cell; right one is the lower half cell.

Figure 10: The CCD pictures of single cell cavity treated by BEP process with the wheel cathode 3. Left one is the upper half cell; right one is the lower half cell.

table 2, although the shapes of thin bar, thick bar and bar cathode seem to have significant differences between each other, the electric field distributions with those cathodes in the cavity do not show appreciable amounts of differences. So, except in the cases of BEP experiments using thin bar cathode, the ratio of removal rate between sample 3 and sample 1 is similar between BEP and EP processes even though there is a relatively great difference in absolute removal rate between BEP and EP processes (see Table 2). It is worth noticing that the electric field distribution we mentioned above is not the one as we mentioned in the etching region. It is the electric field that is rebuilt by an electrochemical polarization process simultaneously at the time of forming the compact solid layer. We think that the formation of oxide layer has two effects: one is that the building of this layer will take appreciable amount of output voltage from the power supply; the other is that the thickness of the oxide layer is different for the different locations on the inner surface of the cavity, leading to a difference in voltage drop at those locations. First effect helps the formation of electropolishing process. The other effect will redistribute the electric field inside the cavity and make it more uniform than that in the etching region.

In the etching region, the electric field has a great difference at different cavity locations due to the geometry of the cavity shape if the normal cathode is thin bar, thick bar or ball. Under such a situation, the field will be much stronger at iris than that at equator. So, if there is no formation of oxide layer at all, the removal rate near iris will be much faster than that near equator. We can see this effect from the data from BEP thin bar experiment as showed in table 2. The removal rate near iris is nearly 5 times larger than that near equator. Due to the much stronger initial electric field at iris, the oxide layer forming at the iris will be either thicker or denser than that at equator. In general, although the effect of the oxide layer cannot make the electric field the same everywhere inside the cavity, the rebuilt electric field distribution will be more uniform than that before. So, in most processes of BEP and EP, the removal rate didn't have a great difference between iris and equator due to the formation of the oxide layer, which is different from the result of the BEP experiment with thin bar cathode as referred above.

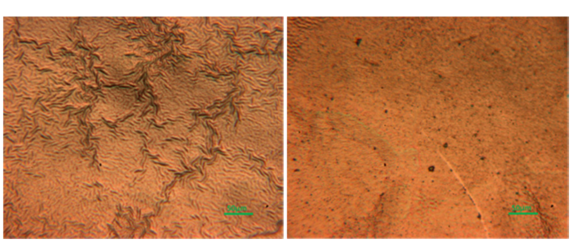

Figure 11: The MOM pictures of button samples from the demountable cavity treated by EP process with the wheel cathode 3. Left one is measured on sample 1; right one is measured on sample 3.

Next, we would like to discuss the reason of the different removal rates between iris and equator. After electric field is rebuilt, the diffusion layer will be formed and becomes the main effect to control the process, leading to a more uniform removal of Nb inside a SRF cavity. Usually, it is thought that diffusion control was not affected by the electric field. However, although we agree with this thought that the diffusion mechanism was independent of the electric field, our experimental results as shown in the following indicated that the boundary conditions of the diffusion layer should be related to the electric field. The location with a larger electric field will increase the number of reactive ions near the boundary of the diffusion layer. According to the Fick's first law, those locations will have a larger current density. Therefore the removal rate of iris will be faster than that of equator since in general the rebuilt electric field after oxide layer formation is still stronger at iris than that at equator. This fact is supported by the results shown in table 2. By using the wheel cathode 3, the ratio of removal rate between equator and iris in BEP and conventional EP became 1.29

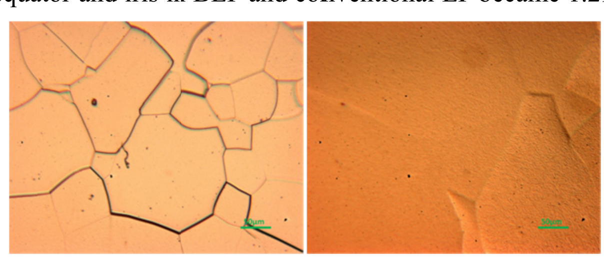

Figure 12: The MOM pictures of button samples from the demountable cavity treated by BEP process with the wheel cathode 3. Left one is measured on sample 1; right one is measured on sample 3.

and 1.16 respectively, since the field distribution is optimized by simulation to be more homogeneous as shown in Fig.6.

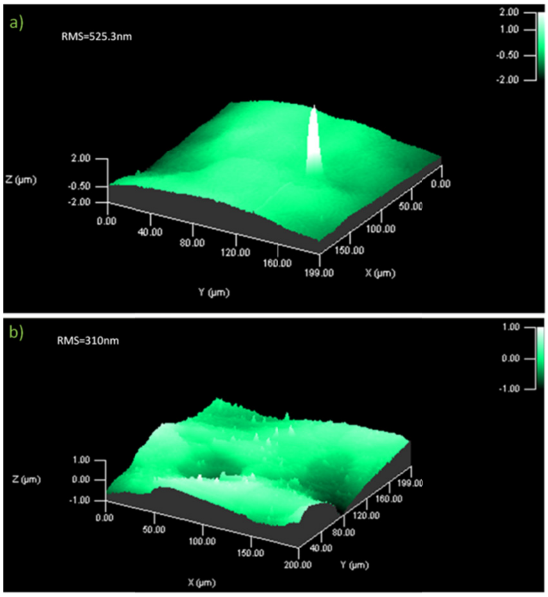

Figure 13: Typical Profilometer images of the surfaces of the button samples from the demountable cavity treated by EP process with a scanned area of (200×200) µm2. a) Sample 1; b) Sample 3.

The Topography of Inner Surface after Polishing

Due to the application of the demountable cavity, the inner surface of the cavity could be directly observed after the polishing processes for both EP and BEP. Figure 9 is the CCD image of an EP treated cavity. The cathode used was wheel cathode 3. Using this cathode shape, the removed thickness of each sample was found to be similar for all area inside the cavity, which made the comparison of the topographic results measured on the button sample 1 with button sample 3 more meaningful. For the same reason, figure 10 is the CCD image of a cathode-wheel-3 treated cavity by BEP process. We can see both of the up-cells of demountable cavity in figures 10 and 11 are shiny and uniform. They should belong to the micro-smoothing processes [3, 4]. The detailed differences were observed and measured by MOM and Profilometer, respectively. Figures 11 and 12 are the MOM images of button samples from EP and BEP processes respectively. The left one is the picture of button sample 1, while the right one is the picture of sample 3 corresponding to each of figures 11 and 12. As we saw, under the resolution of MOM, there

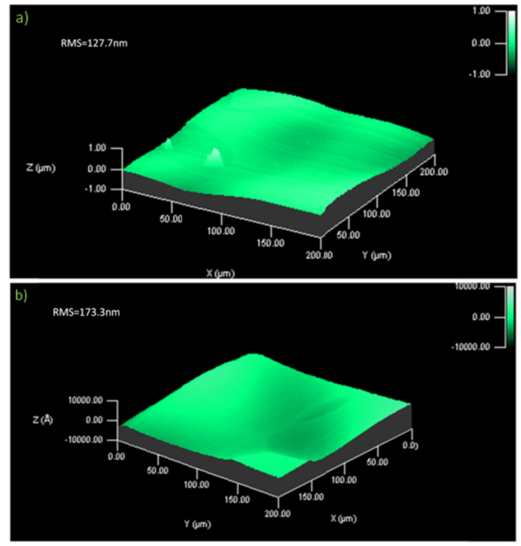

Figure 14: Typical Profilometer images of the surfaces of the button samples from the demountable cavity treated by BEP process with a scanned area of (200×200) µm2. a) Sample 1; b) Sample 3.

are still a little difference between sample 1 and sample 3 for both EP and BEP processes. We think that this difference is understandable due to the effect from the geometry of the cavity shape which cannot allow all the polishing conditions to be established at the same time and the same working point in the I-V curves. So, the subtle difference between iris and equator cannot be completely avoided even if the electric field is optimized.

To see the detailed topography information from the two different polishing processes, the typical profilometer images were obtained with the scanned area of (200×200) µm^2. The samples were also scanned by AFM with (20×20) µm^2 to see possibly more detailed difference. The typical scanned images were shown in figures 13 & 14 and 15 & 16 for profilometer and AFM respectively. From Figures 13a & 13b, we can see the root mean square (RMS) of Fig.13a is larger than that of Fig.13b, implying that iris is rougher during EP treatments. In fact, it is revealed that during EP process, with the wheel shape cathode, the roughness of sample 3 was is always better than that of sample 1 from measurements of both profilometer and AFM. The situation is reversed for the case of BEP as showed in Figures 14a and 14b where it is

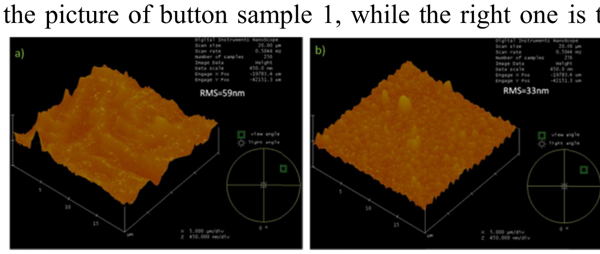

Figure 15: Typical AFM images of the surfaces of the button samples from the demountable cavity treated by EP process with a scanned area of (20×20) µm2. a) Sample 1; b) Sample 3.

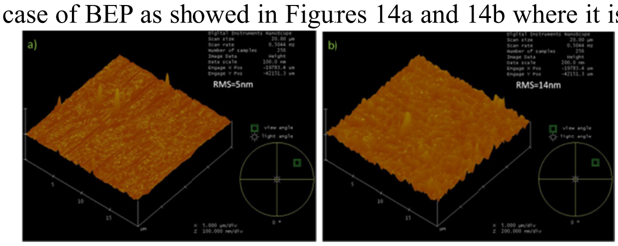

Figure 16: Typical AFM images of the surfaces of the button samples from the demountable cavity treated by BEP process with a scanned area of (200×200) µm2. a) Sample 1; b) Sample 3.

found that iris is a bit smoother. In the BEP experiments, the roughness of sample 1 is always better than that of sample 3. Similar trend is observed by AFM measurements (see Figures 15 & 16). Our experimental results seem to indicate that there is not a direct correlation between the surface roughness and the optimization of the electric field distribution inside the cavity for the EP. For BEP, the correlation somehow exists. A more detailed investigation is underway to clarify the mystery.

To compare the roughness of the button samples during the two different polishing processes, we can see that BEP process always showed a better surface finish than that of the EP via the measurements employed both Profilometer and AFM. Here, we need to notice that the roughness results obtained by Profilometer are quite different from those obtained by AFM. With Profilometer, the RMS of the surface roughness is usually hundreds of nanometers, while the RMS of AFM usually shows tens of nanometers. The surface explored by AFM is so small, that it cannot render the very big defects like e.g. a grain boundary. The roughness detected by AFM is only the intragrain one and does not reflect the large scale roughness, while profilometer probes several grains.

Another thing is worth mentioning here is the difference between upper and lower half cell in the vertical polishing processes. As shown in figures 10 and 11, although the upper half cells are different between the EP and BEP, the lower half cells do not show much difference. Besides, the lower half cells show a lustre surface finish, they are not like a mirror as that of the upper half cells. We think that this should come from the difference in the diffusion layer between the upper and lower cells due to the gravity of the diffusion layer. This difference could have some effects on the RF performance of a Nb SRF cavity. Further research about this topic is underway and the result will be published later.

CONCLUSION

With the help of a demountable cavity, the effects of different cathode shapes on the polishing processes during EP and BEP treatments are studied. Several different cathode shapes such as, for instance, bar, ball, ellipsoid, wheel, etc. were employed. Detailed electropolishing parameters at different locations inside a single cell SRF cavity were measured, including I-V characteristic, removal rate, surface roughness, polishing uniformity and so on. It was demonstrated that optimal polishing results could be achieved by changing the cathode shape for both BEP and EP. The experimental results indicated a close correlation between the electric field inside the cavity and the removal rate. With the optimized wheel cathode via Poisson Superfish simulation, a uniform removal rate was obtained for both EP and BEP processes. It is believed that this fundamental study would provide a useful direction for the development of both BEP and EP for SRF Nb cavity treatments. Through optical CCD images of the whole cavity and the sample analysis by MOM, profilometer and AFM, the roughness of inner surface of the cavity was reported. BEP showed significant advantages in roughness and removal rate in comparison with those of the EP. However, the study also showed the problem about the difference between the upper and lower half cells in the vertical polishing process. This can be one of major problems to be overcome for vertical polishing for both EP and BEP. Further study is on the way and the results will be reported in near further.

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