Development of Niobium Superconducting Cavity

Tomohiro NAGATA[*1], Hirohiko MURAKAMI[*1], Noriyuki ABE[*2], Hiroaki MASUI[*2] and Seiichi SHINOZAWA[*2]

[*1] Future Technology Research Laboratory, ULVAC, Inc., 516 Yokota, Sammu, Chiba, 289-1297, Japan
[*2] Materials Division, ULVAC, Inc., 516 Yokota, Sammu, Chiba, 289-1297, Japan

We study about Niobium refining and elliptical cavity fabrication process for superconducting cavity. In order to carry out Niobium purification, 600 kW electron beam melting furnace was introduced in our factory. It makes possible the stable refining to obtain a cavity quality grade by optimization of melting condition. We performed the trial manufacturing of two single cell cavities are made from our high purity Niobium ingots (RRR>300). Maximum accelerating voltage of welding-type and seamless - type cavities were achieved 41 MV/m and 37 MV/m at 2K, respectively. These values surpass the specification of international linear collider project. Also, seamless tube for three cell cavity was prepared as scale up study. Because an average grain size in the tube for three cell is smaller than that for single cell, it is expected that smoother surface is obtained after hydrofroming process.

1. Introduction

Accelerators are a technology used in a wide variety of fields that have been producing excellent results in research on the origin of the universe, cancer treatment, and analysis of the structure of matter. Any device that can accelerate charged particles can be called an accelerator and there are several methods of accelerating particles, for example, applying voltage electrostatically, using electromagnets, and using high frequency resonance by accelerator cavities, but the most appropriate method is chosen for the application and the type and energy of the particles. Recently, accelerator cavities for which niobium is cooled to the superconducting state are mainly used for MeV to GeV linear accelerators. Large superconducting accelerators are being constructed around the world, such as the European XFEL in Europe, LCLS-II in the U.S., and RISP in South Korea. The International Linear Collider (ILC) project in Japan has been proposed[1]. Niobium materials used in such facilities must be highly pure because they are used in the superconducting state that is sensitive to impurities. The upper limit on the amount of impurities are specified in the ASTM standards as the high purity superconducting grade (ASTM Type 5)[2]. Niobium is rather expensive among pure metals and there are still some problems with technologies for mass-producing accelerator cavities at present, so technologies for manufacturing accelerator cavities at low cost with high productivity are demanded.

The authors have taken a two-pronged approach to developing superconducting accelerator cavities in order to solve the problems mentioned above.

The first is to establish a technology for refining high purity niobium ingots. The goal is to discover a technology for supplying niobium materials that provide the required purity and acceleration performance at the lowest price possible. A 600-kW electron beam melting furnace has been introduced in the factory of ULVAC Tohoku, Inc. to develop methods for manufacturing such niobium ingots.

The second part of our approach is a manufacturing technique called the seamless method. In this method, niobium seamless tubes that are produced by an ULVAC original technology are directly formed into cavities. The aim is to establish a method to manufacture superconducting accelerator cavities at a lower cost than those manufactured by the welding method[3, 4] that is mainly used at present. Some research has suggested that the seamless method in which no welding is required for the main bodies of accelerator cavities has cost advantages[5, 6]. We believe manufacturing costs need to be closely reexamined throughout all the actual processes from purifying raw materials to processing accelerator cavities, as ULVAC has been doing.

This paper introduces ULVAC's two-pronged approach to improve the purity of niobium and to prototype superconducting accelerator cavities by the welding and seamless methods along with their characteristics.

2. Manufacturing high purity niobium ingots

In order to manufacture high purity niobium ingots by electron beam purification, it is best to purify them at high power under high vacuum to effectively remove impurities, especially gases and high melting-point metals. ULVAC has commercialized metal products manufactured by electron beam melting. By using techniques accumulated through such development projects and the advantages we have as a vacuum equipment manufacturer, ULVAC designed a 600-kW electron beam melting furnace for

Figure 1　600 kW electron beam melting furnace.

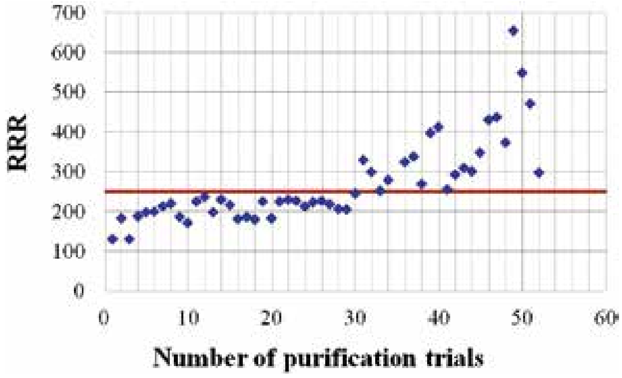

Figure 2　RRR vs. ingot making number.

manufacturing high purity niobium. Figure 1 shows the appearance of the electron beam melting furnace.

After the furnace was first put into operation, melting tests were carried out dozens of times in order to figure out the conditions required for high purification. In this field, the residual resistivity ratio (RRR) is usually used to evaluate purity. This value is often included in specification values when accelerator materials are purchased. The formula below shows the RRR for niobium[7].

$$RRR = \rho\ (300\ K)\ /\rho\ (9.3\ K)$$

This formula shows the ratio of electrical resistivity at 300 K to that at 9.3 K. The superconducting transition temperature of niobium is approximately 9.3 K. At the moment when the temperature falls below this value, the electrical resistance becomes zero. The finite number immediately before superconducting transition is used in the formula. In this state, lattice vibration ceases and so almost no electrons will scatter. Therefore, it can be concluded that electrical resistance is only caused by scattering due to impurities. Thus, the RRR is used as an index for judging the amount of impurities. As the purity gets higher, the RRR value becomes larger.

In the ILC project, the RRR required for niobium materials is more than 250 as specifications. Currently, high purity ingots that satisfy such a value can be produced using this melting furnace (Figure 2). When the furnace was first introduced, its ultimate pressure, electron beam irradiation power, irradiation method to materials, and other matters were being adjusted, so a particularly high RRR could not be achieved. Around the 30th trial, we started to understand the relationship between all the variable parameters and the RRR to some extent, so it started exceeding 250 after the 30th trial and reached as high as 650 under some conditions. Note that it is not that the RRR values varied under the same melting purification conditions (after the 30th time, in particular). The variations in the data are because of differences in the melting conditions depending on the purpose. For example, in one trial we attempted to satisfy the specification value in a shorter amount of time (low cost process) and tried to get the highest purification effect (high RRR) in another. Table 1 shows the chemical analysis results of an ingot with an RRR of 330 as a typical example of high purity. This shows that the ingot is high purity superconducting grade (ASTM Type 5).

3. Mechanical characteristics of high purity niobium materials

Purified ingots are formed into plates and tubes

Table 1　Impurity element analysis of Niobium ingot and ASTM Type 5 specification.

(ppm)

	H	O	N	C	Zr	Fe
ASTM Type5	5	40	30	30	100	50
ULVAC	1	<10	<10	10	<10	<10
	Si	W	Ni	Ti	Al	Ta
ASTM Type5	50	70	30	50	50	1000
ULVAC	<10	10	<10	<5	<10	140

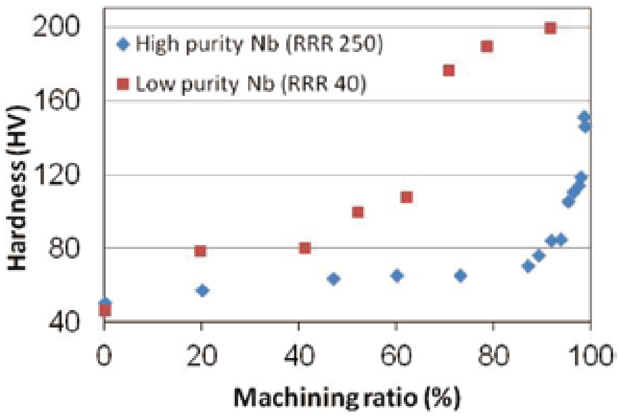

Figure 3　Hardness curve with respect to machining ratio of high purity and low purity Niobium.

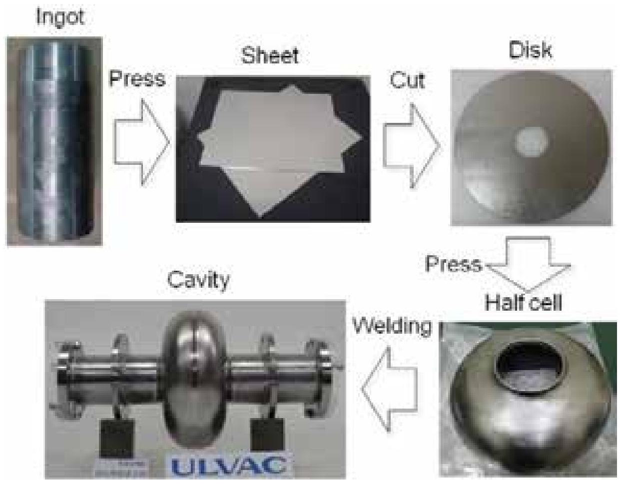

Figure 4　Fabrication process for welding type cavity.

through processing steps. Mechanical characteristics of the materials (e.g., simply whether they are hard or soft) need to be understood to process ingots. Therefore, the degree of processing curve (how the hardness changed when the material was rolled) was created to get an idea of how to machine the metal. The degree of processing refers to a quantitative value that shows how much the material has deformed by processing. For example, if the thickness of a plate is halved, the degree of processing is 50% and if the thickness becomes one tenth, the ratio is approximately 90%.

In this experiment, a high purity ingot (RRR = 250) and a low purity ingot (RRR = 40) were used for comparison. The low purity ingot was a commercially available one with a purity of 3 N. Both types of ingots, which had thicknesses of approximately 10 cm, were repeatedly rolled and the Vickers hardness was measured at different times. Figure 3 shows the degree of processing curve obtained from these measurements. Firstly, for the low purity material, the hardness starts increasing when the degree of processing is approximately 40% and it suddenly jumps up when the ratio exceeds 60%. After that, when the ratio exceeds 80%, the rise of the hardness becomes slow. This is possibly because in the low purity material containing many impurities, as processing proceeds, dislocations get twisted around the impurities and the hardness increases immediately after the processing begins. However, as the material is further processed, it reaches the limits of processing where most dislocations stop moving any further. On the other hand, for the high purity material, work hardening is not seen until the degree of processing becomes relatively high and the hardness finally starts increasing when the ratio exceeds 86%. The limits of processing was not seen even when the material was processed to the degree of processing of 99%. This shows that dislocations do not get entangled much because there are so few impurities in the high purity material. In this experiment, the thickness became 0.3 mm at this stage and further reduction processing became impossible due to the limitations of the rolling machine, so data at higher the degree of processing could not be obtained. The authors will continue to study the relationship between purity and work hardening in the future.

From these results, when low purity niobium materials are heat treated at the degree of processing of approximately 80%, where the materials are close to the limits of processing and strain is sufficiently accumulated, it is possible that recrystallization occurs. On the other hand, not much strain has been accumulated for high purity niobium materials at this stage, so it is probably best to heat treat them after the degree of processing exceeds 95%.

4. Prototyping an accelerator cavity and evaluating its characteristics

4.1 Prototyping a single-cell accelerator cavity using the welding method

A high purity niobium ingot with an RRR of 330 was used to produce a single-cell accelerator cavity using the welding method. The ingot was cut and rolled and the obtained square plates were formed to circular plates as shown in Figure 4. The plates were heat treated in the course of the processes, when required, depending on the accumulated processing strain. The circular plates were formed to a bowl shape called a "half cell" by stamping. The edges of two half cells were placed against each other and welded. Beam pipes and flanges were welded to both ends and the inside was given a mirror finish. This completed the prototyped cavity. The bulged section of an ac-

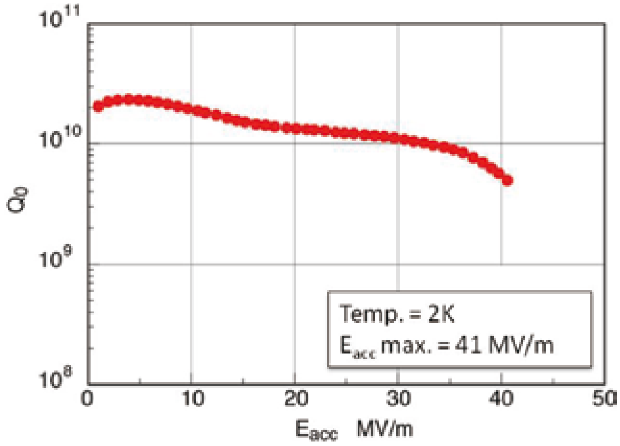

Figure 5 Q_0 - E property of welding type cavity.

Figure 7 Niobium seamless tube for single cell cavity.

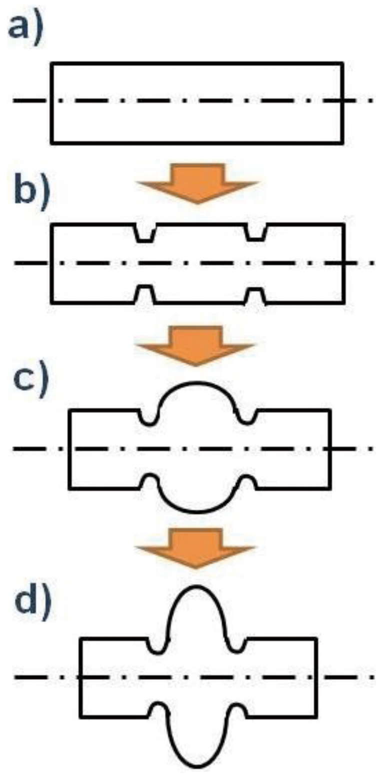

Figure 8 Schematic diagram of manufacturing process of seamless cavity. a) seamless elementary tube, b) after necking, c) after 50% hydroforming, d) after 100% hydroforming.

celerator cavity is called a cell. The prototyped cavity has only one cell, so it is a single-cell cavity. There were concerns that the RRR would decrease as a result of processing and heat treatment. However, the RRR of the circular plate was 324 while the RRR of the original ingot was 330. The difference is within the measurement error, so these two values can be regarded as being on the same level. This shows that processing and heat treatment do not affect the electrical resistivity adversely, in other words, almost no impurities or strain remain. No apparent problems with the welding such as scratches and deformation were seen on the prototyped single-cell accelerator cavity.

This accelerator cavity' s acceleration characteristics were evaluated at the Inter-University Research Institute Corporation High Energy Accelerator Research Organization in Japan (KEK). The accelerator cavity was measured by putting it in a cryostat and liquid helium was used to fill the space between the cryostat and the outside of the accelerator cavity. The cavity was cooled to 2 K. High frequency waves were introduced from the ports at both ends of the accelerator cavity. The ratio of the energy accumulated in the accelerator cavity to the loss of the high frequency waves was calculated to obtain Q_0, which shows the energy efficiency relative to the input electric power. In addition, the net electric field (accelerating electric field, E_{acc}) that particles passing through in the accelerator cavity felt was calculated from the input electric power (for details, refer to documents 8 and 9 in the Refereces). As the input electric power is increased, heat is generated due to the high frequency surface resistance of the accelerator cavity and when the temperature exceeds the superconducting transition temperature, it returns to a normal conducting state (quenching). The accelerating

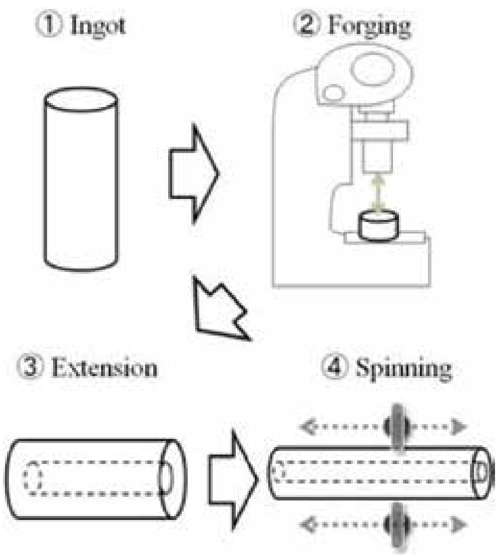

Figure 6 Manufacturing flow of seamless tube.

Figure 9　Single cell seamless cavity.

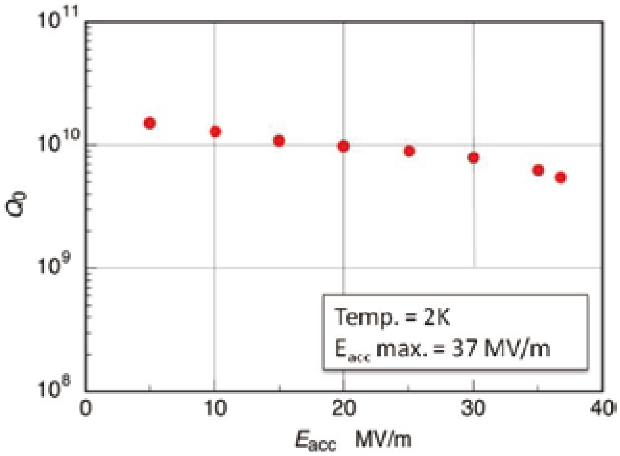

Figure 10　Q_0 - E property of seamless type cavity.

electric field at the time of quenching is called the maximum accelerating electric field (E_{acc}max.). When the input electric power is effectively used as energy to accelerate particles (i.e., when the Q_0 value is higher) and particles can be accelerated to higher energies without quenching (i.e., when the Eaccmax. is higher), the performance of the accelerator cavity is evaluated as excellent.

Q_0 of this single-cell accelerator cavity prototyped by welding was 1.1×10^{10} (when Eacc was 31.5 MV/m) and Eaccmax. was 41 MV/m (Figure 5). These values satisfy the characteristics required for the ILC project ($Q_0 > 1.0 \times 10^{10}$ when Eacc is 31.5 MV/m and E_{acc}max. > 35 MV/m). This demonstrates that ULVAC high purity niobium materials are at a practical level as materials for superconducting accelerator cavities.

4.2 Prototyping a single-cell accelerator cavity using the seamless method

In order to reduce the cost of manufacturing accelerator cavities, ULVAC has been working to use seamless tubes to manufacture accelerator cavities by hydroforming, a method very different from the welding method. To manufacture a seamless tube from an ingot, the ingot is forged, a hole is bored, and then the tube is extended as shown in Figure 6. The tube material was spun to obtain a longer tube while using stronger plastic working. After this process, the tube was ground to adjust the dimensions. The outer diameter of the completed seamless tube for single-cell cavities was 130 mm, the inner diameter was 123 mm, the thickness was 3.5 mm, and the length was 400 mm (Figure 7).

The seamless tube was processed to form a cavity as shown in Figure 8. Both ends of the cell of the original tube (Figure 8(a)) were pushed in to the minimum diameter by necking to form necks (Figure 8(b)). The next process was hydroforming to expand the tube in two steps in which liquid was poured into the tube at a high pressure and then the tube was pressed against dies located outside until it formed the specified shape. The tube was heat treated between the first step (Figure 8(c)) and the second step (Figure 8(d)). The necking and hydroforming were carried out at KEK. The reason why the tube was processed in multiple steps after necking, instead of forming it into the final shape in the hydroforming process in one step, is that the required elongation percentage estimated from the change in the circumference of the equator (the most deformed part) from the shape of the original tube to the final shape is at least 60% (the elongation

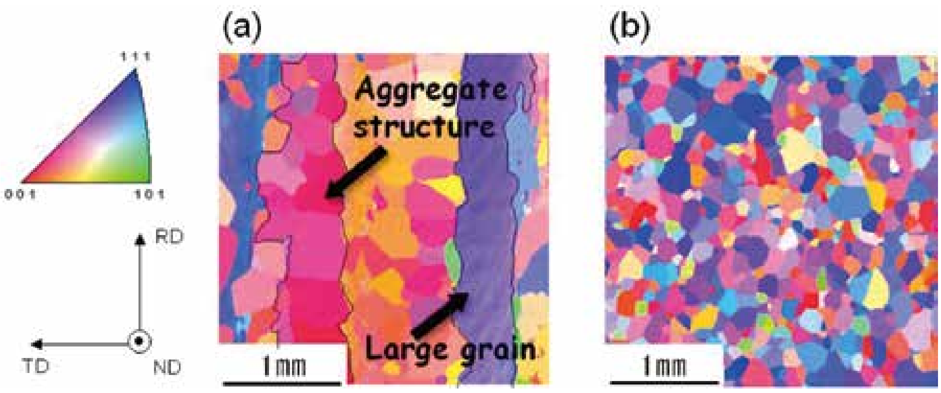

Figure 11　Inverse pole figure (IPF) map of Niobium tube, (a) after weak process, (b) after strong process.

percentage of the seamless tube used was 45%), and it is difficult to obtain such niobium materials. Therefore, heat treatment for crystal recovery is required in the course of hydroforming.

The seamless tube was successfully formed into the cell shape through the processes mentioned above without rupturing. Both ends of the tube were cut off and beam pipes and flanges were welded on. The inside was given a mirror finish. Figure 9 shows the final shape obtained.

The acceleration characteristics of this seamless accelerator cavity were also evaluated at KEK like the welding prototype in the previous section. Q_0 was 8.0×10^9 when E_{acc} was 30 MV/m and E_{acc}max. was 37 MV/m (Figure 10). These values are slightly smaller than those obtained for the welded accelerator cavity. This is possibly because there were some parts where crystal structures were uneven in the seamless tube material and defects like depressions were formed because of differences in the ease with which crystals of different sizes stretched and such differences were exaggerated in the processes involving large displacement, i.e., necking and hydroforming. If such defects could not be completely removed by grinding the inside, it may have disturbed the electric field in the accelerator cavity in the characteristics evaluation, which may have deteriorated the acceleration performance. Specific causes will be studied by conducting inner surface inspections and other inspections. The polishing conditions used in the internal polishing process were determined based on those for the welding method because the technology for the welding method had been established. Therefore, the surface state may not be appropriate for accelerator cavities, so the internal polishing conditions probably need to be optimized too.

Guidelines for material design required for seamless accelerator cavities are explained below based on the prototyping. Cast structures formed during ingot production contain crystal grains of a few cm in some places. These grains are broken by plastic working afterwards. However, it has been proved that if crystal structures are not sufficiently broken, large crystal grains and aggregate structures of crystal grains with similar crystal orientations are formed starting from the remaining grains in the heat treatment after processing[10]. Figure 11 shows differences in crystal structures caused by differences in processing strength observed by the electron back scatter diffraction method (EBSD). In the structure with the estimated degree of processing of less than 70% shown in Figure 11(a), large crystal grains and aggregate structures are seen as indicated by the arrows in the figure. On the other hand, in the structure with the estimated degree of processing of at least 95% in Figure 11(b), the grain size in the structure is rather uniform. When a seamless tube is expanded in the hydroforming process, if the tube has large crystal grains and aggregate structures as shown in Figure 11(a), the elongation of microstructures around them may not follow and then they may break or defects may be formed, which may deteriorate the acceleration performance as this study showed. Therefore, it is desired that the cast structures of seamless tubes that can withstand hydroforming be properly broken and that the uniformity of the grain sizes of crystal structures be high for the entire tube. To that end, the amount of processing by forging and tube expansion, and controlling crystal growth by determining the conditions for heat treatment carried out after processing are the most likely key points.

Figure 12　Niobium seamless tube for three cell cavity.

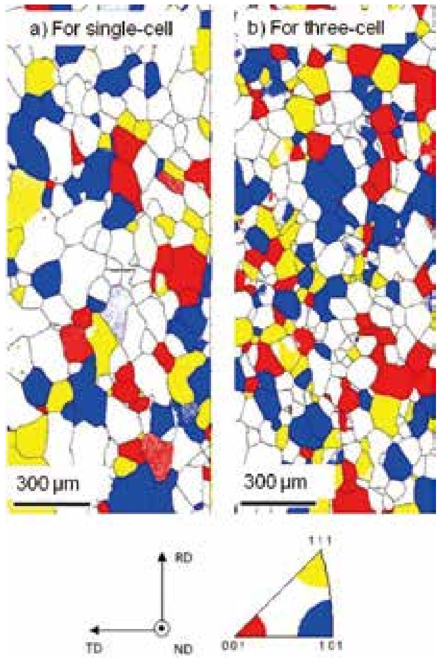

Figure 13　Crystalline orientation of seamless tube for a) single cell cavity and b) three cell cavity.

5. Prototyping seamless tubes for three-cell accelerator cavities

As explained above, the authors have succeeded in forming seamless single-cell accelerator cavities. However, a standard accelerator cavity that is to actually be used in the ILC project and European XFEL project has nine cells (nine-cell type). Therefore, the authors started prototyping seamless tubes to forming three-cell cavities as the next step in the development of seamless accelerator cavities. The first step is producing a nine-cell accelerator cavity by connecting three three-cells. The next step is producing a nine-cell accelerator cavity from a single seamless tube.

A seamless tube was produced in the flow shown in Figure 6 as was the tube for single-cell cavities in the previous section. A seamless tube for three-cell cavities was prototyped through the processes mentioned above (Figure 12). The outer diameter was 138 mm, the inner dimeter was 131 mm, the thickness was 3.5 mm, and the length was 830 mm. One difference with the seamless tube for single-cell cavities was that the diameter of the tube was increased by 8 mm. This was to reduce the amount of expansion needed to reach the final diameter from the original diameter in the hydroforming to avoid the risk of the tube rupturing in the process. Another difference was that the number of rollers in the spinning machine was increased from two to three. This modification made it possible to transmit the force from the rollers to the tube more evenly, which improved the processing dimension accuracy. The tube for single-cell cavities was machined to the target dimensions after the spinning process. The modification to the rollers made it easy to form cavities with dimensions within the target tolerances upon completion of the spinning process. Omission of the grinding process improved the yield of materials by 25%. The prototyped seamless tube is currently being prepared for hydroforming.

Evaluation results of a test piece cut from the seamless tube are explained below. First, the RRR was 350 and no degradation was seen after it was processed. As for its mechanical characteristics, the tensile strength was 155 MPa and the elongation percentage was 53%. We know that these values compare favorably with those of existing high purity niobium materials for cavities. As mentioned above, when tubes are formed into seamless accelerator cavities, controlling crystal structures is very important to reducing the risk of rupture. Crystal structures in the seamless tube for three-cell cavities were compared to those of the seamless tube for single-cell cavities described in the previous section by using EBSD. Figure 13 shows the observation results. More smaller crystal grains are seen in the tube for three-cell cavities. The average diameter of crystal grains in the tube for single-cell cavities is 121 μm. On the other hand, it is 89 μm for the tube for three-cell cavities. This is probably because stronger processing was able to be applied to the material thanks to the modification to the spinning machine. Regarding the crystal orientations, the crystal grains for which the incline is within 15 degrees from (001), (101), or (111) are illustrated using the same color for each orientation. If aggregate structures exist, they are seen as a string of crystals of the same color as shown in Figure 11(a). However, potential aggregate structures are not seen for both types, which shows that the crystals are rather randomly oriented. From these results, it can be said that the tube can withstand deformation by hydroforming as is the case with the tube for single-cell cavities and that the surface roughness should be able to be reduced because the average diameter of crystal grains in the tube for three-cell cavities is smaller. The authors will make more prototypes and evaluate them to quantitatively evaluate what crystal structures are most desirable for hydroforming.

6. Conclusion

This paper explained the development of materials through a series of processes from purification of niobium ingots to prototyping of accelerator cavities. A high-power electron beam melting furnace was introduced to manufacture ingots. The authors have demonstrated that high purity ingots with an RRR of more than 250 that is required for general materials for superconducting accelerator cavities can be manufactured. ULVAC will develop technologies focusing on throughput and cost reduction by closely inspecting raw materials and melting purification conditions. Such high purity niobium ingots made by ULVAC were used to prototype single-cell accelerator cavities using the welding and seamless methods. The performance of the prototyped cavities was relatively good as both types satisfied the acceleration characteristic ($E_{acc\text{-}max.} \geq 35$ MV/m) specified in the ILC project. However, the Q_0 value of the seamless accelerator cavity needs to be improved. ULVAC will work to improve the value by controlling crystal structures and optimizing the polishing conditions. In addition, seamless tubes for three-cell cavities were prototyped as a higher-level development of the seamless accelerator cavities. The results indicate that strong processing was applied which reduces the risk of rupture during hydroforming thanks to the modification to the processing equipment. ULVAC will aim to produce seamless nine-cell accelerator cavities and verify their cost advantages in comparison with the welding method.

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ULVAC, Inc. 2500 Hagisono, Chigasaki, Kanagawa, 253-8543 Japan TEL :+81-467-89-2033 www.ulvac.co.jp/en/