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A NOVEL MANUFACTURE OF NIOBIUM SRF CAVITIES BY COLD SPRAY

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Abstract

Niobium coating technology has been developed for superconducting radiofrequency (SRF) cavities to reduce material costs. Cold spray (CS), a film formation method that feeds solid-state particles onto a target to form a thick film in a short time, is the focus of this study, with the aim of reducing the cost. To verify the feasibility of manufacturing an entire structure of SRF cavities using CS, an additive lathe method with CS was performed. Niobium particles were supplied through a CS gun manipulated by a robot arm to consistently orient the blast direction to the normal direction of an aluminum alloy sacrificial mandrel. Nb-made cavities were successfully fabricated by dissolving the mandrel in hydrochloric acid.

INTRODUCTION

Superconducting radiofrequency (SRF) cavities are a key component of particle accelerators, and they are mainly made of niobium (Nb) because they possess the highest transition temperature (T_c) of all pure materials [1, 2]. There are higher-T_c compounds such as NbN and Nb_3Sn, but at present, they do not match Nb in terms of their performance with increasing RF fields or ease of use for accelerator applications. In other words, Nb is still the best option for SRF cavities [2]. However, Nb is a rare metal and its material cost is a problem. Thus, Nb coating on copper (Nb/Cu; hereafter, a layer structure consisting of the outermost layer X_1, second layer X_2, third layer X_3, …, and substrate Y is denoted as $X_1/X_2/X_3.../Y$) technology has been researched to reduce the raw material cost [3]. Additionally, Nb/Cu technology provides more advantages, such as thermal stability, insensitivity to earth magnetic field trapping, and freedom from undissolved inclusions [2, 3]. Sputter coating was developed at CERN for Nb/Cu coating in 1980 [4], and research has been conducted to overcome problems such as the theoretical limit of bulk Nb and the *Q*-slope [5]. As a unique alternative, Fonnesu et al. [6] developed a reverse coating that employed a sacrificial Al mandrel to form a Cu/Nb/Al structure, which was subsequently etched to obtain the Cu/Nb structure. They also reported that the Cu/Nb/Cu/Al structure was better for the Nb layer because the Cu layer between Nb and Al protected Nb from the final etching.

Because the London penetration depth λ_L of Nb is as short as ~40 nm and the SRF properties are determined only on the surface, a couple of micrometers is sufficient for the thickness of the deposition film in theory [2]; however, in practice, the thickness affects the technical difficulties of the pre- and post-process. For example, the surface roughness prior to a sputtering process strongly influences the quality of the deposition film [3]; thus, a mirror finish of several nanometers is required on the entire internal surface of the cavity. In addition, a shallow deposition thickness does not allow post-processing to correct the shape defects of the film.

Cold spray (CS) is a film formation method; different from conventional thermal spraying techniques, CS feeds solid-state particles with supersonic speed to form thick films on the target [7]. Because the particles remain in a solid state throughout the process, CS can suppress oxidation, thermal stress, and high temperatures [8]. Additionally, the main advantage of CS over other film formation methods for SRF cavities, such as high-power impulse magnetron sputtering (HiPIMS) [5] and electron cyclotron resonance (ECR) [9], is its deposition rate; in our preliminary research, the deposition rate of the CS reached 300 mm^3/min, which was several digits greater than that of the sputter method.

Kumar et al. [10] reported that CS successfully deposited a superconducting Nb film on a Cu substrate and confirmed that the film transited to the superconducting state. CS was also applied to form a thick copper layer on Nb SRF cavities, hoping to enhance the conduction cooling ability [11]. However, the fabrication of the entire structure of Nb SRF cavities has yet to be reported. Therefore, we attempted to manufacture models of seamless SRF cavities using CS to confirm their feasibility. In addition, superconducting tests were performed to confirm the superconducting performances of the test pieces.

EXPERIMENTAL

Cold Spray

The niobium powder employed in the CS experiment was manufactured by Taniobis GmbH, Germany (Ampertec® TS niobium metal powder 10–30 µm), and its microphotophy is illustrated in Fig. 1. The chemical compositions are listed in Table 1.

Cold spray was performed using a commercially available device (PCS-1000, Plasma Giken Co., Ltd., Japan [12]), and it was manipulated by a robot arm with six degrees of freedom (Motoman MS80, Yaskawa Electric Corp., Japan). The experimental conditions for the CS

Table 1: Chemical Compositions of the Niobium Powder Measured by Manufacturer (unit: wt. ppm)

C	H	N	O	Fe	Cr	Ni	Mn
<10	<10	57	526	<2	<2	<2	<1
Na	K	Mg	Ca	Si	Mo	Ta	W
<1	<1	97	<3	<3	<4	73	<5

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process are listed in Table 2. Note that a preliminary test under a gas temperature of 1000 °C generated many spatter sparks of Nb particles, which may cause degradation of the Nb film. Therefore, the lower temperature of 450 °C was chosen for this experiment.

The samples for the superconducting tests were fabricated from a cold-sprayed (CSed) Nb film on an oxygen-free copper plate (JIS C1020 grade). The toughness and thermal conductivity of aluminum are advantageous for minimizing the deformation of the substrate owing to mechanical and thermal stresses.

As the structure of the SRF cavities is rotationally symmetric, the additive lathe method was adopted to form a thick film on the outer surface of the mandrel, as illustrated in Fig. 2. Nb powder was supplied by the spray gun during rotation to deposit the outer surface of the mandrel. The position and attitude of the spray gun were manipulated using a robot arm to consistently orient the spraying direction toward the normal direction of the surface of the mandrel. The mandrel was made of JIS A2024, which is equivalent to the 2024 aluminum alloy. Its shape was similar to 3.9 GHz SRF cavities with dimensions of 90 mm (longitude) × 76 mm (equator). It was clamped with screw-ended shafts and washers that prevented adhesion were inserted between them. They were then clamped by a chuck and rotated by the spindle.

Superconducting Test and Hardness Test

The CSed Nb film was removed from the Cu plate and sliced into 1 × 2 × 100 mm test pieces using a wire electric discharge machine (WEDM). Hereafter, these test pieces are called 'As-received' samples. Their electrical resistance was tested in a cryogenic vacuum chamber with four-terminal sensing to confirm the superconducting transition. The samples were cooled to ~5 K by conduction cooling, and the temperature was gradually increased after the chiller was turned off. The samples were measured again after performing a standard treatment for the Nb SRF cavities: buffered chemical polishing (BCP) with a mixture of hydrofluoric acid, phosphoric acid, and nitric acid, and vacuum annealing for 3 h at 800 °C. Additionally, the hardness of the as-received and annealed samples was tested using a Vickers hardness tester (HM-100, Mitutoyo Corp., Japan) with a load of 1 kgf (9.80665 N).

RESULTS AND DISCUSSION

Figure 3 illustrates the progress of the CS process on an Al-alloy mandrel. Figure 3(b) shows that an Nb film started to form even in the first scan, and a thick Nb film can be seen in Fig. 3(c). Figure 4 depicts the cross-sections of (a) the Nb/Al mandrel sliced by WEDM, (b) Nb thick film after dissolving Al mandrel in a 35 wt% HCl solution, and (c) Nb thick film rinsed in the BCP solution. Figure 4 confirms that the CS process enables the deposition of a seamless over 2-mm Nb film on the Al mandrel within a machine working time of 320 s. It should be noted that the Nb film was discolored in copper-like red after HCl

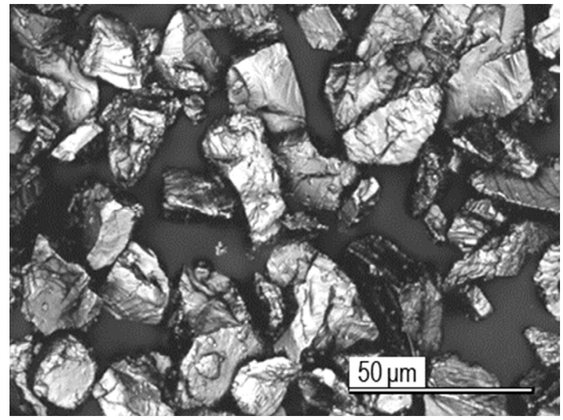

Figure 1: Laser microscopic photo of niobium powder observed with a confocal laser scanning microscope (VK-8510, Keyence, Japan).

Table 2: Conditions of Cold Spray for (a) Plate and (b) Mandrel.

	(a)	(b)
Working gas	Nitrogen	
Gas temperature	450 °C	
Gas pressure	3 MPa	
Nozzle traverse speed	20 mm/s	~ 20 mm/s
Lathe rotation	—	250 min^{-1}
Standoff distance	20 mm	30 mm
Number of repetitions	10	9
Substrate material	Cu[*1]	Al alloy[*2]

*1: Oxygen-free copper (JIS C1020), *2: JIS A2024

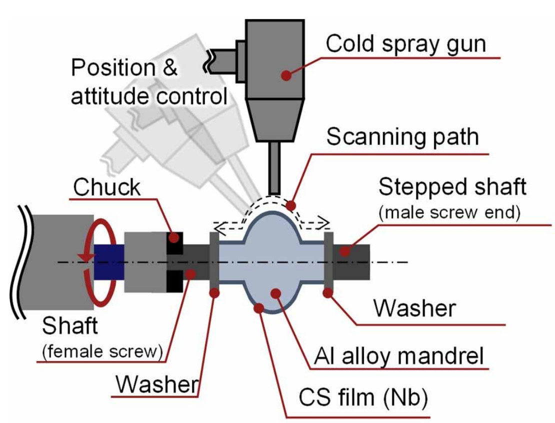

Figure 2: Schematic of the CS process for fabricating a cavity model.

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dissolution, indicating that copper in A2024 of the mandrel material might appear as shown in Fig. 4(b). The subsequent BCP process turned the color into gray, but the test piece did not exhibit metallic luster as Fig. 4(c). Its density was measured through Archimedes' principle and was 7.0 g/cm^3, smaller than 8.56 g/cm^3 of bulk Nb.

Compared with previous studies [6, 10] performed on flat plates, this result guarantees the applicability of the proposed method to a three-dimensional structure.

Figure 5 shows the relationship between the temperature around the superconducting transition temperature (T_c) and the resistivity of the CSed samples, indicating that a superconducting transition appeared on both samples. At present, it is unclear what causes the difference in the transition temperatures between the samples, and further research is required. Table 3 lists the residual resistivity ratio (RRR) and hardness values of the as-received and annealed samples. The RRR value was comparable to previous research, 3.42 [10], and other coating methods, 10–26 with magnetron sputtering [14] and 4.36 HiPIMS [15]. It decreased after BCP and annealing, which is consistent with previous research [10]. This phenomenon may be caused by the CS process, but further research is required to clarify this. The hardness of the as-received samples was more than double that of a bulk Nb sheet (50 HV in [16]), and the hardness of the annealed samples decreased. Such hardening is typical of CSed coatings owing to plastic diffusion [17], and heat treatment is reported to be effective in relaxing residual stress [18].

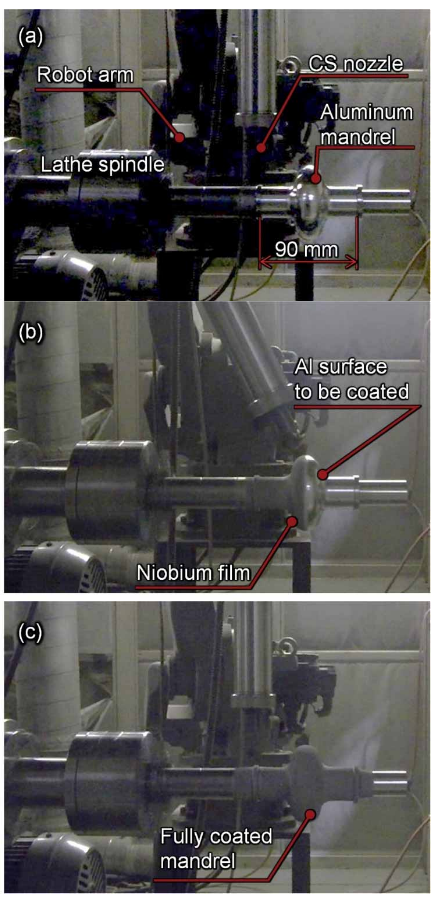

Figure 3: Photographs of the progress of the CS process: (a) before CS, $t = 0$ s, (b) in the middle of the first pass $t = 18$ s, and (c) after CS, $t = 320$ s, where t is the elapsed time of the CS process.

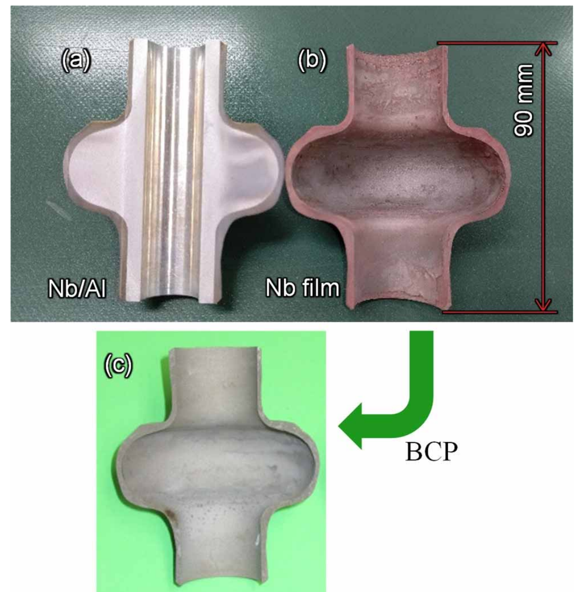

Figure 4: Cross-section of the Nb/Al mandrel similar to 3.9 Hz SRF cavities: (a) as-CSed Nb/Al mandrel, (b) Nb film alone dissolved by HCl solution, and (c) Nb film through BCP solution.

Table 3: RRR and Hardness (mean values and standard deviations in parentheses)

Samples	As-received	Annealed
RRR*	11 (0.1)	8 (0.1)
Hardness (HV1)	116 (3)	101 (11)

RRR = R(293 K)/R(Tc), where Tc* is the temperature just above the transition temperature [13].

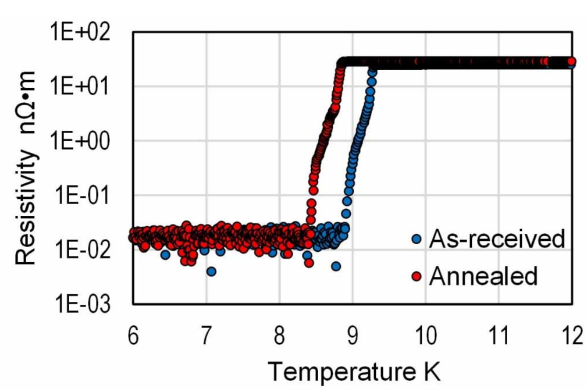

Figure 5: Temperature–resistivity diagrams of as-received and annealed samples.

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CONCLUSIONS

Cold spray was first applied to manufacture an entire model of the SRF cavities. An Nb-fabricated structure was successfully fabricated on an aluminum alloy mandrel. These RRR values were comparable to those reported in previous studies. The next step in this feasibility study is to manufacture a 1.3 GHz cavity to evaluate the radio-frequency performance.

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