Silicon Nitride Spraying Using Quasi-Steady MPD Arcjets

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Abstract: In magneto-plasma-dynamic (MPD) arcjet generators, plasma is accelerated by electromagnetic body forces. Silicon nitride reactive spraying was carried out using an MPD arcjet generator with crystal silicon rods and nitrogen gas. Because higher-velocity, higher-temperature and higher-density and larger-area plasmas are produced with the MPD arcjet generator than those with conventional thermal plasma torches, nitriding of silicon can be enhanced. A dense and uniform β-Si_3N_4 coating 30 μm thick was formed after 200 shots at a repetitive frequency of 0.03 Hz with a discharge current of 9 kA and a substrate temperature of 700 °C. The Vickers hardness reached about 1300. Velocity and temperature histories of silicon particles generated in the MPD generator nozzle were numerically simulated. The velocity and temperature of particles increased downstream. The velocity reached about 1750 m/s at the nozzle exit, and the particle was completely melted when the particle diameter was 1 μm. This result shows that the MPD arcjet generator has high potentials for silicon nitride spraying.

I. Introduction

The quasi-steady magneto-plasma-dynamic (MPD) arcjet generator is a promising plasma accelerator, which has a coaxial electrode structure similar to those of conventional plasma torches.[1] However, their acceleration mechanisms are different; i.e., the MPD arcjet generator utilizes principally electromagnetic acceleration of the interaction between the discharge current of kiloamperes and the magnetic field azimuthally induced by the discharge current, although the working gas is accelerated aerodynamically through a straight or convergent-divergent nozzle in a thermal plasma jet generator. In megawatt-class input-power repetitive pulsed operations, higher-velocity, higher-temperature, higher-density and larger-area plasmas can be produced with the MPD arcjet generators than those with conventional plasma sources.[2,3] The discharge plasmas are expected to be utilized for various material manufacturing processes.[4-8]

In the present study, silicon nitride reactive spraying is carried out using an MPD arcjet generator with crystal silicon rods and nitrogen gas. The coating properties are analyzed with SEM and XRD. Vickers hardness is also measured. Furthermore, velocity and temperature histories of silicon particles generated in the MPD generator nozzle, which affect coating properties, are calculated computationally.

II. Experimental Apparatus

Figure 1 shows the cross-sectional view of the coaxial MPD arcjet generator developed for silicon nitride spraying. The rod cathode 6 mm in diameter is made of thoriated tungsten. The anode nozzle made of copper has an exit diameter of 58 mm and a half angle of 20 degree. Four crystal silicon rods (cross section: 5 mm x 1 mm; purity: 99.999%) are located in the divergent nozzle, and the distance between the edge of the silicon rods and the cathode tip is 20 mm. Nitrogen is used as the working gas. As shown in Fig.2, a high-current arc is expected to melt the silicon rod surface, and then the ablated silicon particles react with nitrogen plasma.

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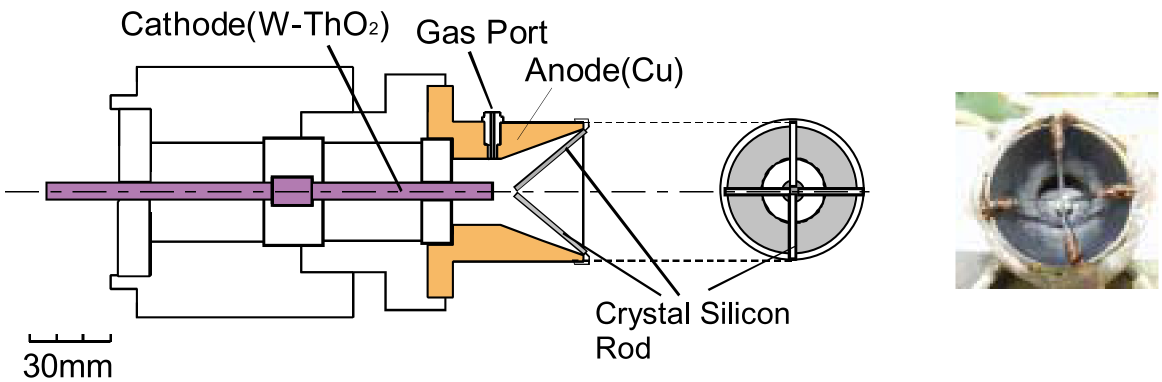


Figure 1. Cross-sectional view of MPD arcjet generator for silicon nitride reactive spraying.

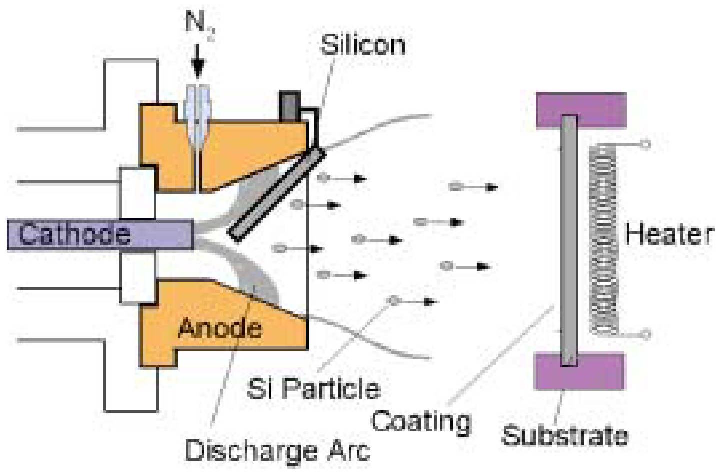


Figure 2. Illustration of silicon nitride reactive spraying using MPD arcjet generator.

The main power-supplying pulse-forming network is capable of storing 62 kJ at 8 kV and delivers a single nonreversing maximum quasi-steady current of 27 kA with a pulse width of 0.58 ms. The MPD arcjet generator is installed on a stand in a vacuum tank 1 m in diameter x 1.2 m in length. The tank pressure is kept at $2\text{-}5\times10^{-2}$ Pa during periodical operations. Unprepared substrate plates 125 mm x 125 mm x 4 mm, made of stainless steel SS400, are placed at 150 mm downstream from the arcjet generator exit. The substrate temperature is controlled ranging from room temperature to 700 ^{o}C with an electrical heater behind the substrate plate.

III. Silicon Particle - Plasma Flow Interaction Calculation

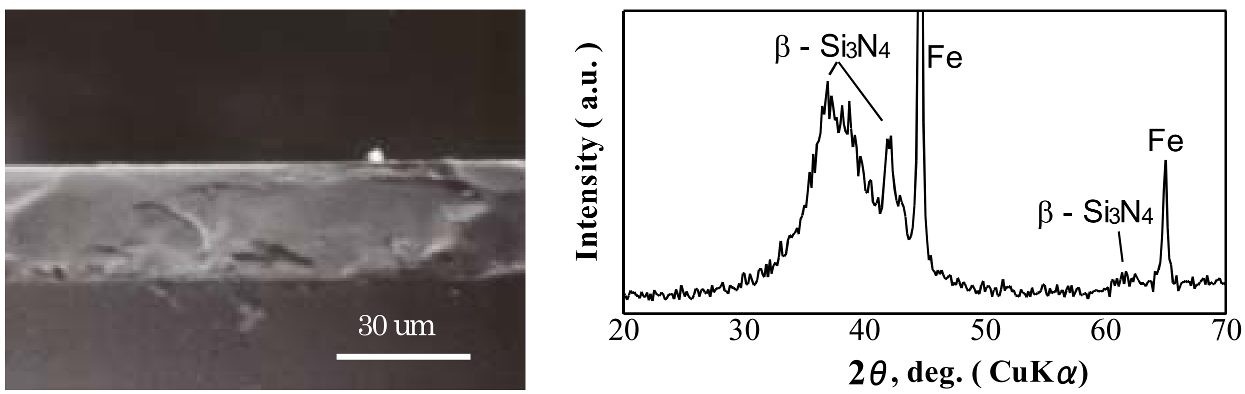
Triple-probe measurements were made to evaluate electron temperatures and electron number densities of the MPD arc plasmas, and emission spectroscopic measurements were also conducted to identify excited ion and atom species in the plasmas.[9] Axisymmetric MPD generator flowfield was computationally calculated in order to understand the nitrogen plasma acceleration processes.[10,11] Conservation equations of mass, momentum and energy in addition to the magnetic field equation are a group of modified Euler equations. By using all plasma diagnostic and flowfield calculated data, velocity and temperature histories of silicon particles generated in the MPD generator nozzle, which affect coating properties, are calculated.

In this calculation model, a silicon particle with an axial velocity of zero is injected at 20 mm downstream from the cathode tip on the central axis. The silicon particle is assumed to be axially accelerated by the drag force from the plasma flow and to be uniformly heated by energy transfer from the plasma flow. Two models of momentum and heat transfers are expressed as a function of the Knudsen number.[12,13]

IV. Results and Discussion

A. Coating Characteristics

The MPD arcjet generator for silicon nitride reactive spraying was operated with 200 shots at a repetitive frequency of 0.03 Hz (1shot/30sec) for a N_2 mass flow rate of 2.5 g/s. Figure 3 shows a photograph of cross section and the XRD pattern of the coating with a substrate temperature of 700 °C at a discharge current of 9 kA. A dense and uniform coating 30 μm thick is formed after 200 shots. If the MPD arcjet generator is operated at a repetitive frequency of 0.05 Hz (1shot/20sec), the deposition rate is expected about 10 times higher than those for conventional CVD and PVD methods. As shown in Fig.3(b), peaks of Si are not identified, and the microstructure of β-Si_3N_4 is created. Figure 4 shows the thickness of the coating dependent on discharge current. The thickness rapidly increases from 5 μm at 6 kA to 30 μm at 9 kA. This is expected because an amount of silicon particles ablated by current concentration on the crystal silicon rods intensively increases as the discharge current increases.



(a) Cross-sectional view **(b) XPD pattern**

Figure 3. Photograph of cross section and XRD pattern for silicon nitride coating after 200 shots with N_2 mass flow rate of 2.5 g/s at discharge current of 9 kA and substrate temperature of 700 °C.

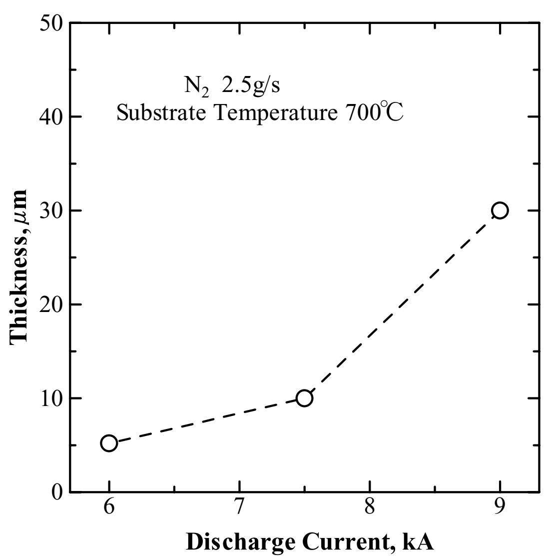


Figure 4. Thickness vs discharge current characteristics for silicon nitride coating after 200 shots with N_2 mass flow rate of 2.5 g/s at substrate temperature of 700 °C.

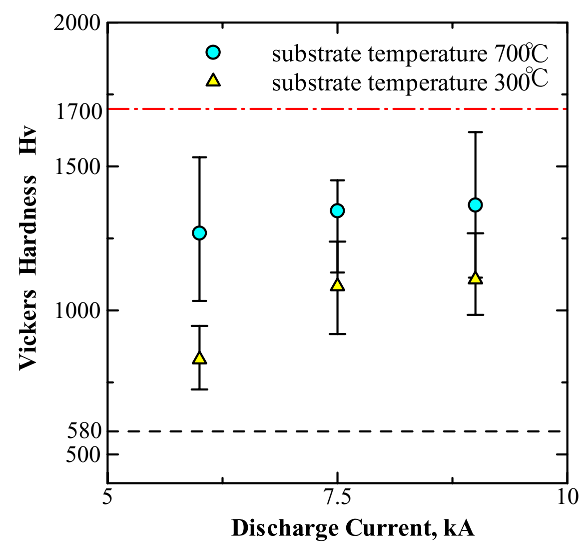
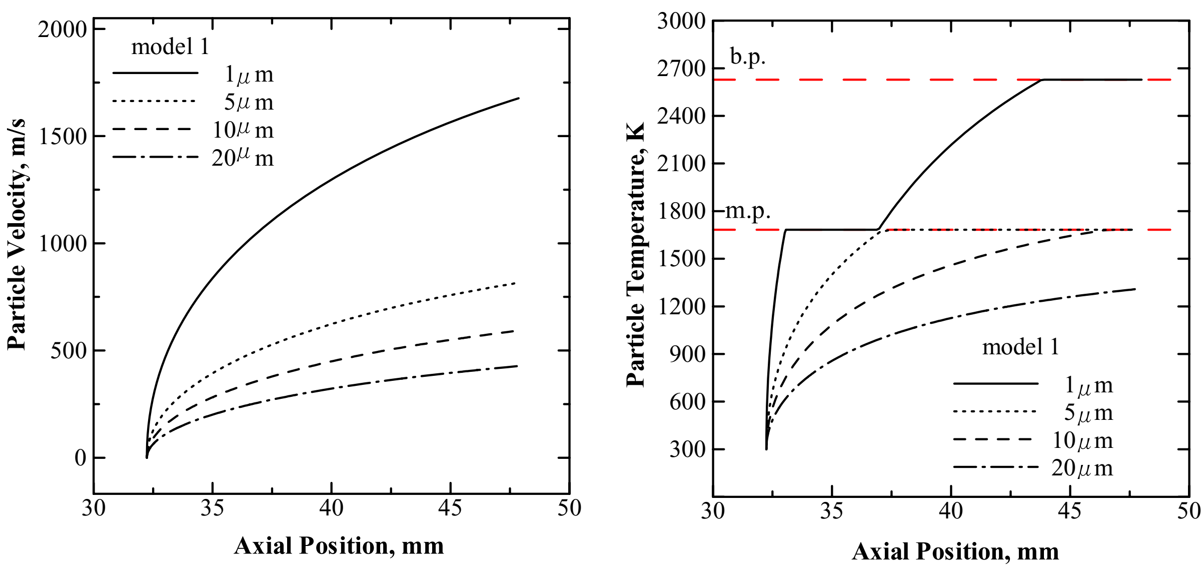


Figure 5. Vickers hardness vs discharge current characteristics for silicon nitride coating after 200 shots with N_2 mass flow rate of 2.5 g/s at substrate temperatures of 300 and 700 °C.

Figure 5 shows the Vickers hardness of the coating at substrate temperatures of 300 and 700 °C dependent on discharge current. The Vickers hardness increases from 1250 at 6 kA to 1300 at 9 kA with 700 °C and from 800 at 6 kA to 1100 at 9 kA with 300 °C. More adhesive coatings are deposited with increasing discharge current because of particles sprayed with higher-velocity and higher-temperature. At a constant discharge current, the hardness at 700 °C is higher than that at 300 °C. In XRD patterns, as the substrate temperature decreased, the β-Si_3N_4 peak became broad. Sprayed particles are rapidly solidified on the substrate as the substrate temperature decreases. Therefore, the microstructure of the coating is changed from crystal to amorphous, resulting in decreasing hardness. Accordingly, very hard coatings are deposited although depending on discharge current and substrate temperature.

B. Velocity and Temperature Characteristics of Sprayed Silicon Particles

The velocity and temperature histories, i.e., in-flight characteristics, of small silicon particles ablated from the crystal silicon rods are roughly estimated. Figure 6 shows the velocity and temperature histories of silicon particles through the divergent nozzle at a discharge current of 6 kA dependent on particle diameter. The calculated results with model 1 almost equaled those with model 2.[12,13] The particle velocity gradually increases downstream. At a constant axial position, a decrease in particle diameter raises the velocity. The particle velocity with 1 μm reaches about 1750 m/s at the nozzle exit. In particle temperature, as shown in Fig.6(b), the temperature rapidly increases downstream. The particle temperatures for small particles of 1, 5 and 10 μm reach the melting temperature in the divergent nozzle, and furthermore the temperature with 1 μm reaches the boiling one. Accordingly, high-velocity and completely-melted sprayed particles are expected to be expanded from the nozzle exit of the MPD arcjet generator.



(a) Particle velocity **(b) Particle temperature**

Figure 6. Calculated velocity and temperature histories of silicon particles through divergent nozzle at discharge current of 6 kA dependent on particle diameter.

V. Conclusions

Silicon nitride reactive spraying was carried out using an MPD arcjet generator with crystal silicon rods and nitrogen gas. A dense and uniform β-Si_3N_4 coating 30 μm thick was formed after 200 shots at a repetitive frequency of 0.03 Hz with a discharge current of 9 kA and a substrate temperature of 700 °C. The Vickers hardness reached about 1300. It was found that coating thickness was highly sensitive to discharge current and that microstructure of the coating was sensitive to substrate temperature. Velocity and temperature histories of silicon particles generated in the MPD generator nozzle, which affect coating properties, were numerically simulated. The velocity and temperature of particles increased downstream. The velocity reached about 1750 m/s at the nozzle exit, and the particle was completely melted when the particle diameter was 1 μm. This result shows that the MPD arcjet generator has high potentials for silicon nitride spraying.

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