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Citation	Transactions of JWRI. 35(1) p.29-p.33
Issue Date	2006-07
oaire:version	VoR
URL	https://doi.org/10.18910/10357
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Gun Current Optimization for Preparation of Silicon Carbide Films by Gas Tunnel Type Plasma Spraying†

FAHIM F. Narges* and KOBAYASHI Akira**

Abstract

Polycrystalline silicon carbide films have been prepared by the gas tunnel type plasma spraying method (GTPS). The effect of gun current on microstructure and mechanical properties was investigated. Scanning electron microscopy, x-ray diffraction, energy dispersive spectroscopy, nanoindentation and abrasive wear were used to characterize the structure, thickness, composition and the mechanical properties of SiC films. Microstructural studies revealed the formation of cubic silicon carbide (C-SiC) at higher gun currents from 120 to 140 A. The SiC films have good-adhesion, dense, smooth and compact morphology. Determination of hardness of the SiC films by a nanoindentation technique shows that increasing gun current can improve hardness from 25.3 to 31.5 GPa. Generally, SiC film formed at higher gun currents exhibits better anti- wear resistance than that deposited at low gun current, mainly due to SiC films becoming harder. A comparison of the hardness of SiC films grown by gas tunnel type plasma spraying and SiC films grown by other methods was included. Finally, crystalline silicon carbide films with good morphology and mechanical properties have been obtained from the GTPS method that are suitable for thermoelectric and mechanical applications.

KEYWORDS: (Silicon carbide), (thermoelectric materials), (Microstructure), (Hardness), (Wear behaviors), (Gas tunnel type plasma spraying).

1. Introduction

Silicon carbide is an important structural material because of its unique combination of properties, such as high resistance to wear, corrosion, and thermal shock, chemical inertness, high temperature strength, high stiffness, high thermal conductivity, high breakdown electric field and mechanical and electrical stability at high temperature. Hard materials like silicon carbide have received great attention for many applications, as aeronautic, high-temperature (> 600 ^{o}C) MEMS sensors (pressure, temperature, gas and optical) microprocessors, memory devices and power-generation devices. Material properties, like high hardness, low friction and low wear versus metals are essential for this purpose. Although, diamond like carbon is fulfill these requirements but they have limited applications as their microstructure, hence tribological and mechanical properties are strongly changed at high temperature. Doping DLCs with elements such as silicon (up to 30 at. %) should extend their applications to high temperature environments (T > 675 K) [1].

A variety of techniques are used to grow single and polycrystalline forms of SiC thin films, including atmospheric pressure chemical vapor deposition (APCVD), sputtering, metal-organic CVD, atomic layer epitaxy, molecular beam epitaxy, and pulsed laser deposition (PLD). Each technique has its own merits and limitations. The main drawbacks of CVD are slowness, relatively high expense, need for high temperature and contamination of the films [2-4].

Plasma spraying is an economical and convenient method for preparation of refractory materials. The high melting point of SiC ~ 2700 ^{o}C requires the plasma spray instrument to run at high power. High power gas tunnel type plasma spraying (GTPS) has been developed [5, 6] and is used to achieve efficient melting and good quality films. Comparison with conventional plasma spray technique, high power gas tunnel type plasma spraying can produce materials with 20-30 % higher hardness and density. Moreover, GTPS system is simple, relatively inexpensive and requires much lower substrate temperatures than CVD. Deposition of several ceramic materials by gas tunnel type plasma spraying has been previously reported [7-9].

In this study, silicon carbide films have been prepared by gas tunnel type plasma spraying. The effect of gun current on microstructure and mechanical properties was investigated. The microstructure features of SiC were analyzed using scanning electron microscopy (SEM), energy dispersive electron spectroscopy (EDS) and x-ray diffraction (XRD). While, mechanical properties were determined by measuring the micro-hardness and abrasion wear resistance.

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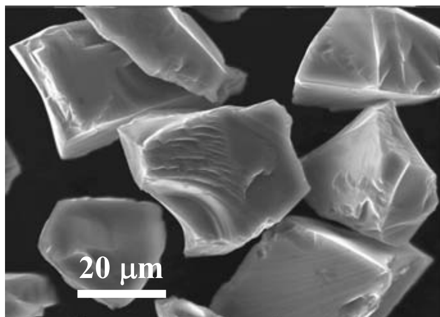
Transactions of JWRI is published by Joining and Welding Research Institute, Osaka University, Ibaraki, Osaka 567-0047, Japan

2. Experimental Procedure

2.1 Preparation of SiC films

High purity SiC powder, moissanite-2H, of particle size 20-45 µm was atmospherically plasma sprayed (APS) by using a gas tunnel type plasma jet. The morphology and phase structure of the powders are shown in **Fig. 1**. The chemical composition and purity of the used SiC powder are given in **Table 1**. The used substrate material was AISI 304 stainless steel with dimensions ($50x50x3mm^3$). Before spraying deposition, the substrate was grit blasted using alumina powders to clean and roughen the surface.

SiC films were synthesized using a gas tunnel type plasma spray torch, which has been previously described [10]. The torch was composed of two anodes and one cathode and two power sources. The external nozzle diameter is 20 mm and the internal one is 8 mm. The flame diameter is almost 20 mm to melt the majority of injected powder with high efficiency. SiC powder was internally feed inside the plasma flame stream to obtain maximum flame temperature because SiC powder has a high melting point ≈ 2700 ^oC. The spraying deposition of silicon carbide was performed under various gun currents and spraying parameters were listed in **Table 2**. The deposition efficiency of the spray process and the characteristics of deposited films are strongly influenced by the properties of the starting powders. Powders flowability and sprayability are among the most important parameters. Flowability is depends on the spraying gun and the nozzle. Sprayability is strictly correlated to the kinetic and thermal behavior of the single powder granules: too large particles may require too high dwell times within the plasma to attain a uniform temperature and be efficiently molten; small, light particles, on the other hand, can never enter the main stream of the plasma jet, remaining solid at the periphery of the jet and including debris and porosity within the deposit. The particles shape, size and size distribution are the main control parameters.



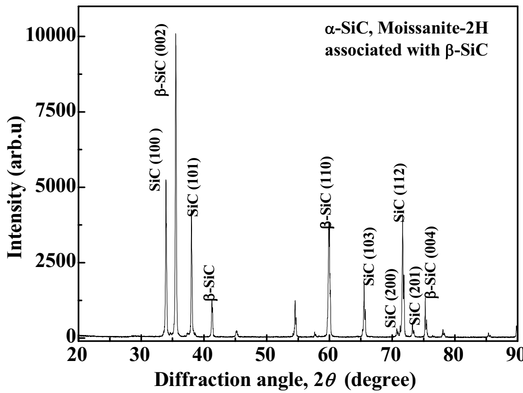


Fig. 1 SEM micrograph and XRD diffraction pattern of feedstock SiC powder.

Table 1 The chemical composition and the purity of the silicon carbide powder

α-SiC (moissanite- 2H)	Weight percentage (%)
SiC purity	> 97
C	28-30
Si	67-69.5
[O]	2-2.6

Table 2 Spraying parameters used for the preparation of SiC films.

Gun current [A]	80, 100,120, 140
DC plasma vortex current [A]	450
Primary gas flow rate, Q [Ar, l/min]	150
Carrier gas[Argon, l/min]	10
Powder feed rate, w [g/min]	5
Spraying distance, L [mm]	50
Spraying time, t [sec]	20
Nozzle diameter [mm]	20
Traverse rate [mm/s]	nil

2.2. Film characterization

2.2.1. Microstructural investigation

The microstructure and morphology of the SiC powder and deposited SiC films were observed by a scanning electron microscope (SEM, ERA 8800FE). The chemical composition of the SiC films was investigated with a scanning electron microscope equipped with an energy dispersive spectrometer (EDS) detector. The phases of the SiC feedstock powder and SiC films were identified by X- ray diffractometer (XRD-Cu-Kα , JEOL JDX-3530M).

2.2.2. Mechanical characterization

(a) Micro-hardness determination

A Vicker's indenter was used to measure the hardness using the Akashi AAV-500 series microhardness tester. The Vicker's microhardness measurements were done on polished and buffed surfaces with load: 200g, load time: 20 s and T= 25 ^oC. This load was used for deposited SiC films while a load of 25 g was used for SiC powder, due to the high hardness and brittleness of SiC powder. At least ten indentations were made for each load to provide

satisfactory statistics. The hardness Hv in GPa was calculated from the following equations:

$$Hv = 1852.f.d^{-2} \quad (1)$$

Where f is the load (N), d is the average length of the diagonals of the Vickers indents (mm).

(b) Abrasive wear test

The abrasive wear tests were conducted on SiC films in air without lubricant using a SUGA ABRASION TESTER which follows the NUS-ISO-3 standard. **Figure 2** shows a systematic diagram of the wear tester apparatus. It consists of movable wheel driven by an electric motor under load. The specimens were fixed on movable plate and moved horizontally (oscillation mode) a distance of 1cm under load 25 g on a slightly moved wheel (10 mm/min) covered with strip of SiC paper. The wide of the wheel is 1 cm and the contacted area with the specimen is 1cm^2. The specimens were exposed to move against the abrasive wheel at time intervals (5-20 s). Then the sample was continually examined for the same time range (5-20 s.) For all specimens the weight loss was measured before and after load time using a 6 digit electronic balance.

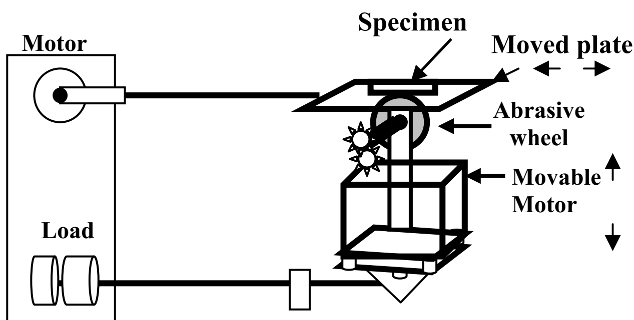


Fig. 2. Systematic diagram of abrasive wear tester.

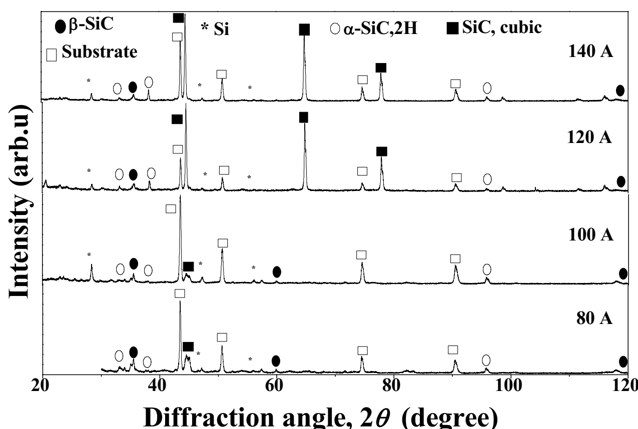


Fig. 3 XRD patterns of SiC films prepared under various gun current of 80, 100,120, and 140 A.

3. Results and Discussions

3.1. Microstructure

The morphology and microstructure of feedstock SiC powder were depicted in Fig.1. The peaks corresponding to α-SiC (100), (101), (103), (200), (201), and (112) planes were clearly presented. Moreover, the (002), (110), and (004) peaks due to the presence of β-SiC, have been detected. Therefore, the starting powder is α-SiC of hexagonal crystal structure.

Figure 3 shows the XRD patterns of SiC films deposited under various gun currents of 80, 100, 120, and 140 A. Obviously, as the gun current increase, a new phase of SiC started to appear. The new peaks of (200), (220) and (311) planes are related to cubic silicon carbide (C-SiC), which clearly appeared at high gun currents of 120, 140 A. While the peaks intensity correspond to α-SiC of hexagonal structure (H-SiC) started to decrease as the gun current increased. Also, peaks due to the presence of silicon of planes (111), (220), and (311) have been seen. These peaks could be attributed to the decomposition of some SiC particles, in the region of high temperature of the flame, into Si and C. Finally, other peaks can be seen in the XRD patterns that are related to the stainless steel substrate of planes (111), (200), (220), and (311). The presence of substrate peaks in the XRD patterns of SiC films was attributed to the thickness of the film, which ranged from 5 to 12 μm.

Scanning electron micrographs of SiC films sprayed under different gun current of 80 and 140 A are shown in F**ig. 4**. The morphology of SiC films prepared at a gun current of 140 A are depicted in fig.4a,b. Clearly, the SiC films are smooth, compact, dense and well-adhered. It is seen that the only difference between SiC films deposited at gun current of 140 A compared with films grown at 80 A relates to the thickness and the smoothness. Mostly, SiC films grown at 140 A have thickness of ~ 12 μm and more smooth than those deposited at low gun current. This is mainly due to the increase in the total power of the plasma spray system, which consequently increases the flame temperature that causes melting the majority of SiC particles.

3.2. Mechanical properties

3.2.1. Hardness

Indentation testing is critical to the understanding of mechanical properties of films. The indentation test is strongly affected by the surface roughness of the film. In general, very smooth surfaces are required to obtain reliable contact stresses at small depths of indentation. Indentation of films is also dependent on the penetration depth due to the underlying substrate effects and hence study of indentations with contact depths of less than 10–20 % of the film thickness is recommended to ensure the intrinsic properties of the film (to reduce the substrate interference). **Figure 5** shows the Vicker's microhardness of SiC films obtained at various gun current of 80, 100, 120 and 140 A. The hardness increased with increasing

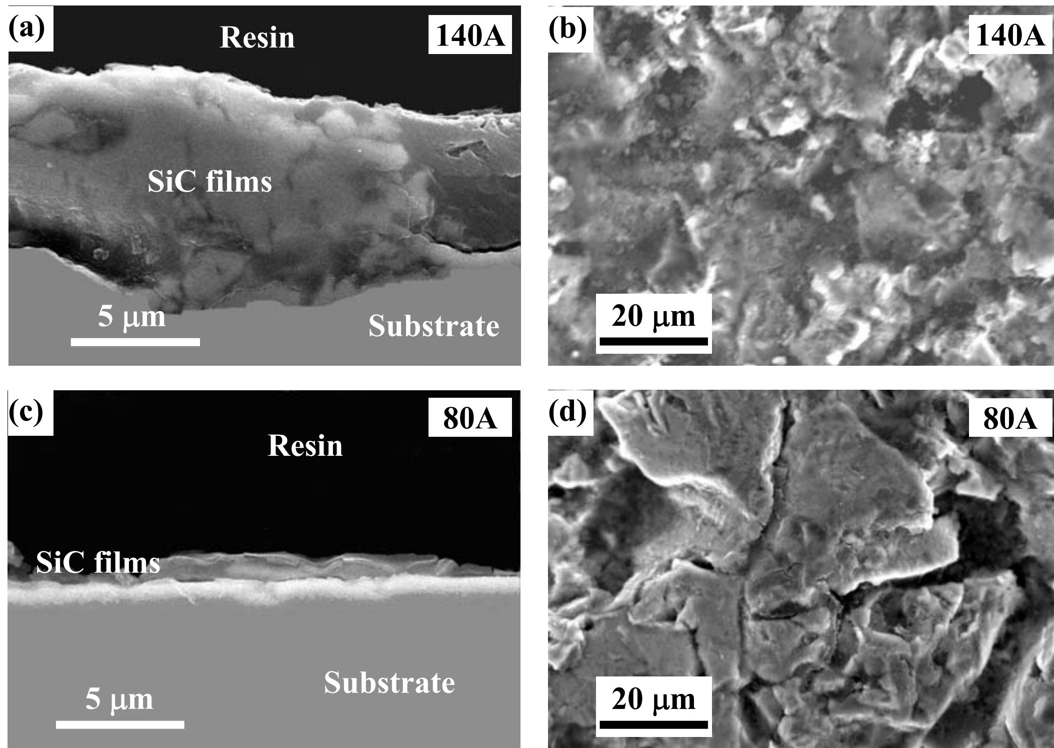


Fig.4. Scanning electron micrographs of SiC films sprayed under different gun current: (a) 140 A, cross-section view , (b) 140 A, surface-view, (c) 80 A, cross-section view and (d) 80 A, surface –view.

the gun current from 80 to 140 A. The hardness was about 25.3 GPa for the film grown at a gun current of 80 A and increased to approximately 31.6 GPa for the film deposited at 140 A gun current. These hardness values are in good agreement fwith crystalline cubic-SiC grown by PACVD. The experimental results show the hardness decreased with increasing penetration depth, as is typical for behavior of a hard film on a soft substrate. If the film is harder than the substrate, then most of the plastic deformation occurs in the soft substrate, leading to a ''sink-in'' effect even though the indenter is not yet through the film. The variation of hardness with depth of indentation for GTPS and CVD films are quite the opposite, possibly due to the presence of impurities on the surface in CVD film.

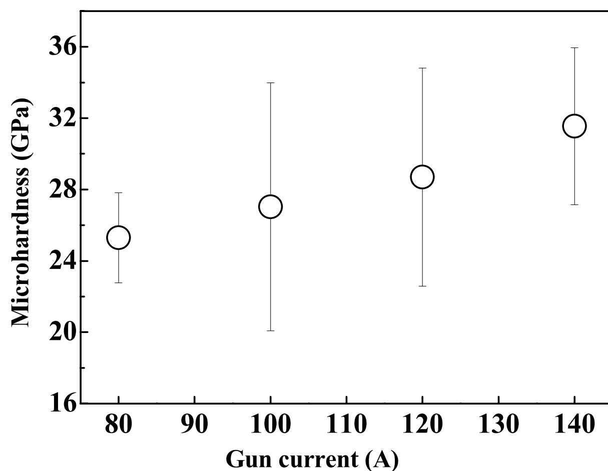


Fig. 5 Variation of hardness of SiC films as a function of gun current.

Table 3 shows the comparison between hardness of SiC films grown by different methods such as PACVD, CVD, sintering, ns-PLD and fs-PLD. The differences in hardness are primarily attributable to the type of structure, amorphous or crystalline, and to the degree of Si-C covalent bonding.

Table 3 Comparison between hardness of SiC films grown by gas tunnel plasma spraying and other methods.

Preparation method	Hardness (GPa)
PACVD [*11*]	21-30.4
CVD [*12*]	45
Sintering, SSiC [*13*]	31.3
ns-PLD [*14*]	22-36
fs-PLD [**12**]	10-20
IBAD [**15**]	22
GTPS [***this work***]	25.3-31.6

3.2.2. Wear behaviors

In order to observe if such films are of interest for mechanical applications, wear under load was tested. The abrasive wear resistance of SiC plasma deposited under different gun currents is shown in **Fig. 6**. The results indicated that the films obtained at high gun current exhibit higher abrasion resistance compared with those formed at lower gun currents. The abrasive wear rate of SiC films at 140 A is 3 x 10^{-5} g/cm^2/min, which is lower than value of abrasive wear rate of SiC formed at 80 A (3.3 x10^{-5} g/cm^2/min). Apparently, wear rates for all films decrease with increasing gun current. This indicates that the wear resistance of the SiC films is significantly improved, as gun current increases. This is mainly due to, on the one hand, SiC films becoming more dense and hard, as the gun current increases, which act as a barrier for wear. On the other hand, the adhesive bond between the SiC particles becomes stronger due to the increase in the total power of the plasma system. Therefore, the corresponding SiC films formed by GTPS would be good candidates for low friction under high loads.

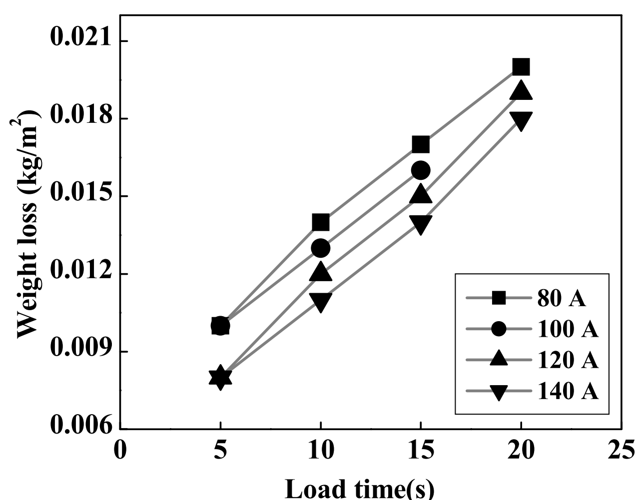


Fig.6 Abrasive wear rate of SiC films deposited under different gun current.

4. Conclusions

This work is focused on the preparation of SiC films in the gas tunnel type plasma device. In this process, experimental parameters are chosen to check the influence of gun current on the structure and mechanical properties. The main conclusions are as follows:

(1) The microstructure results showed that polycrystalline cubic-SiC are obtained in high gun current from 120-140 A.
(2) SEM studies indicated extremely compact, dense, well-adhere and smooth surface for films of approximately 12 μm thickness,, for a film grown at 120-140 A.
(3) Cubic- silicon carbide films formed by gas tunnel type plasma spraying have proper hardness (25.3 – 31.5 GPa) compared to the hardness values in the literature.
(4) SiC films grown at higher gun current exhibit better wear-resistance than that deposited at low gun current.

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