

Preparation of SiC Matrix Laminated Ceramic Composites by Electrophoretic Deposition

Yang Zhou, Akiko Honda,* Tomokazu Takeda,* Masato Uehara,* Naoya Enomoto* and Junichi Hojo*

National Key Laboratory of Advanced Composites, Beijing Institute of Aeronautical Materials, Beijing 100095, China
Fax: 86-10-6245-8002, e-mail: yangzhou66@hotmail.com

*Department of Applied Chemistry, Faculty of Engineering, Kyushu University, 6-10-1 Hakozaki, Higashi-ku, Fukuoka, 812-8581, Japan
Fax: 81-92-642-3547, e-mail: jhojotcf@mbox.nc.kyushu-u.ac.jp

Preparation technique of silicon carbide/carbon laminated composite by electrophoretic deposition was developed in this investigation. Both acetone and deionized water media were used for the deposition of SiC layers. Deposition rate of SiC powder from acetone-base suspension was far higher than that from aqueous suspension, indicating that the acetone medium is more suitable for the deposition of thick SiC layers. Variation of deposition weight as a function of deposition time and applied voltage was also studied in acetone suspensions with solid concentration of 10 wt% and 15 wt% for SiC, and 7.5 wt% for carbon black. The laminated deposit was shaped by alternate deposition from SiC suspension and carbon black suspension. After the deposit was uniaxially pressed and then dried, the laminated composite was obtained by hot pressing. Typical laminated structure with the layer thickness of about 75 μm for SiC matrix layer and about 8 μm for carbon interlayer was observed by SEM. Load-displacement response of the laminated composite exhibited non-catastrophic fracture process when the composite was tested by three-point bending method.

Key words: laminated composite, electrophoretic deposition, silicon carbide, carbon

1. INTRODUCTION

Some nature materials, such as marine shells, possess hierarchical structure formed by aragonite layers that are joined by a mortar of protein [1]. The bending strength and toughness of these materials increase by one order of magnitude compared to a single crystal of aragonite because of this configuration [2]. In the last decade, ceramic matrix composites with similar laminated structure have been developed and considered to offer one of the most important approaches to the problem that ceramic materials lack in damage tolerance [3-5]. It has been known that the brittleness of ceramics can be improved by introducing weak interlayers, which may deflect a growing crack, and thus, prevent catastrophic failure. High toughness, work-of-fracture and reliability were exhibited in laminated ceramic composite systems due to the presence of weak interlayers [6-9].

Various techniques, such as tape casting [10], slip casting [11], gel casting [12] and centrifugal casting [13] etc, have been used to prepare laminated ceramic composites. Electrophoretic deposition (EPD) technique is also a facile forming method demonstrated in recent years for the synthesis of laminated composites [14]. EPD has good shaping capability, and the shape is formed in a continuous way. Moreover, various kinds of particles can be distributed homogeneously in a deposit by EPD when they are mixed and deposited simultaneously. By alternately depositing in two suspensions of different ceramic compounds, EPD allows the formation and shaping of the laminated

composites in one step process. The deposition rate and weight are controllable, and the uniform layers with the minimum thickness of about 2 μm in laminated composites have been attained by EPD technique [15]. Such a thin layer is very hard to be made by other method. Many examples of materials, that have been deposited electrophoretically, demonstrate that the process is widely applicable. In the present study, SiC matrix laminated ceramic composite was shaped successfully by EPD technique, and non-catastrophic fracture characteristic was shown for the laminated composite.

2. EXPERIMENTAL PROCEDURE

The raw materials and solvents used in this investigation were commercially supplied. The matrix layer of laminated composite was composed of fine SiC powder (Ibiden Co., Ltd, Japan). Al_2O_3 and Y_2O_3 powders (99.9%, Rare Metallic Co., Ltd, Japan) were used as sintering additives. The chemical composition, mean particle size and specific surface area (SSA) of the SiC powder are given in Table I. The interlayer of laminated composite consisted of carbon black powder (2300[#], Mitsubishi Chemical Corporation, Japan).

To compare the EPD behavior of SiC powder in different media, both water and acetone were used to prepare the matrix suspensions. Aqueous suspension with 10 wt% solid concentration in deionized water was ultrasonically agitated for 20 min and followed by magnetic stirring for 2 h. The pH value of the suspension

Table I. Chemical composition (wt %), mean particle size and specific surface area of SiC powder[†]

SiC	f-C	f-SiO ₂	T-Al	T-Fe	H ₂ O	Mean size, μm	SSA, m^2/g
98.4	0.85	0.40	0.02	0.05	0.31	0.28	23.6

[†] As given by the supplier.

was adjusted to 10 by adding ammonia. Acetone suspensions with 10 wt% and 15 wt% solid concentrations were prepared by the similar process with addition of 10 vol% n-butylamine. The suspension of 7.5 wt% carbon black was also prepared by the same way in acetone medium with 10 vol% n-butylamine for the deposition of laminated composite interlayers.

The cell used for electrophoretic deposition comprised a graphite disk with a diameter of 15 mm as deposition electrode and a stainless steel sheet as counter electrode. The deposition voltage was controlled with a variable DC power supply. The deposition took place on the graphite electrode by controlling its positive polarity of the applied voltage. The suspension was stirred gently and continuously with a magnetic stirrer to prevent the powder from settling during deposition. For deposition rate test, SiC deposits were formed by applying the electric field of 5V, 7.5V, 10V and 12.5V for different time intervals of 5min, 10min, 15min and 20min with an electrode distance of 10mm. The carbon black powder was deposited under the same electric fields but different time intervals of 1min, 2min, 3min and 4min. The deposit was weighed after dried, and the same weight of new powder was added to the suspension to keep the concentration constant.

The laminated composite was shaped by alternate deposition from matrix suspension and carbon black suspension with the acetone medium including n-butylamine. The sintering additives of 5 wt% Al₂O₃ and 3 wt% Y₂O₃ for SiC were mixed in the powder suspension. Since the deposition rate decreased with the increase in deposit thickness, the applied voltage was increased stepwise from 8 V by 0.2 V interval after stacking each SiC layer. Different deposition times, i.e. 8 min for each matrix layer and 1.5 min for each carbon black layer, were used in the EPD process. A binder of 1 wt% polyvinylbutyral was added to both the two suspensions to increase the green strength of the deposits. After depositing the required number of SiC and carbon black layers, the deposit was removed from the graphite electrode, and then uniaxially pressed in the wet state using a graphite mold to prevent the boundary between matrix layer and interlayer from cracking. The composite was sintered by hot pressing (HP) at 2000°C and 50 MPa for 30 min under 1 atm Ar atmosphere.

A bar specimen with a size of 15 mm \times 2.5 mm \times 2 mm was cut from the laminated composite block, and three-point bending test was carried out with a universal testing machine (AG-5000D, Shimadzu Co., Japan). The span size was 12 mm and the loading rate was 0.5 mm/min. Morphologies of the laminated composite were observed with a scanning electron microscope (SEM, S-2300, Hitachi Co., Japan) before and after sintering.

3. RESULTS AND DISCUSSION

3.1 EPD behavior of SiC powder in different media

Two kinds of media, i.e. deionized water with ammonia and acetone with n-butylamine, were used for the deposition of SiC powder. Fig.1 shows the deposit

weights per unit area achieved at applied voltage of 5 V for 20 min with the electrode distance of 10 mm from the two suspensions with 10 wt% solid concentrations. It can be seen that the deposit weight of SiC in acetone suspension is about 18 times larger than that in aqueous suspension. To improve the dispersibility of SiC powder in aqueous suspension, various pretreatments such as acid solution leaching, alkaline solution leaching and calcination in air were carried out for the SiC powder. The deposit weight of SiC did increase obviously after treatment (which will be discussed elsewhere), but it was also far less than that in acetone suspension. Therefore, acetone with n-butylamine is the suitable medium for depositing thick SiC layers.

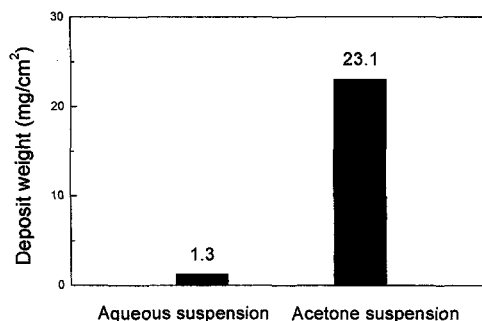


Fig.1 Electrophoretic deposition weights of SiC powder from different suspensions

3.2 Effect of processing parameter on EPD in acetone suspension

The effects of various processing parameters, such as applied voltage, deposition time and solid concentration, on the electrophoretic deposition weights of SiC and carbon black powders were studied in acetone-base suspensions. Fig.2 shows the variation of EPD weight of SiC as a function of applied voltage for deposition time 10 min. As shown, the deposit weight increased linearly with the increase in applied voltage in 10 wt% and 15 wt% SiC suspension. Furthermore, the slope of weight-voltage line, which represents the dependence of deposit weight on applied voltage, increased with the increase in solid concentration. To compare the deposition efficiency of SiC in acetone-base suspensions with different solid concentrations, the deposition weight was normalized by the applied voltage and deposition time. The deposition efficiency, which means the deposition weight per unit area, per unit voltage and per unit time, was obtained from Fig.2 as following: 0.30 $\text{mg}/\text{cm}^2\text{Vmin}$ for 10 wt% SiC suspension and 0.51 $\text{mg}/\text{cm}^2\text{Vmin}$ for 15 wt% SiC suspension.

The effect of deposition time on the SiC yield at applied voltage 10 V is shown in Fig.3. For a certain solid concentration, the deposition weight increased linearly with deposition time. The deposition rate, i.e. the slope of weight-time line, also increased with the increase in solid loading. Therefore, a large solid concentration is

more effective to get a thick SiC layer. The deposition efficiencies calculated from Fig.3 are 0.28 mg/cm²Vmin for 10 wt% SiC suspension and 0.50 mg/cm²Vmin for 15 wt% SiC suspension, which are quite consistent with the results from Fig.2. This is the result from the linear dependency of deposit weight on applied voltage and deposition time within the present conditions.

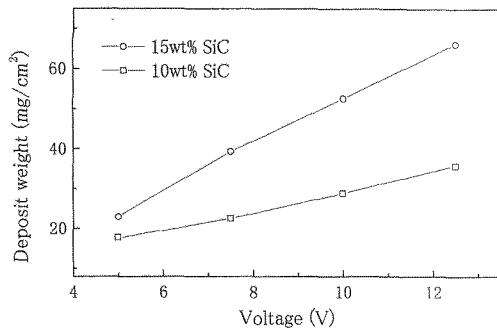


Fig.2 Variation of EPD weight of SiC as a function of applied voltage (Deposition time 10 min).

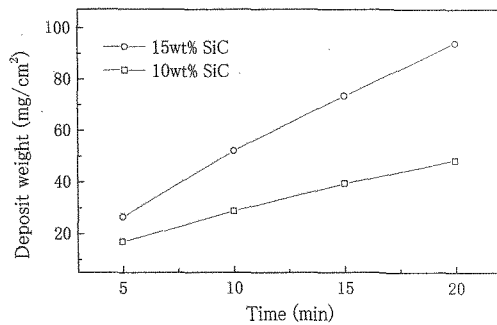


Fig.3 Variation of EPD weight of SiC as a function of deposition time (Applied voltage 10V).

The influences of deposition time and applied voltage on the EPD weights of SiC in 15 wt% suspension and carbon black in 7.5 wt% suspension are shown in Fig.4 and Fig.5, respectively. The results demonstrate that the deposit weight can be easily controlled by deposition time and applied voltage in both powders. The deposition time was long for SiC forming matrix layers of the laminated composite, and short for carbon black forming the thin interlayers. For 15 wt% SiC suspension, the average deposition efficiency counted from Fig.4 was 0.50 mg/cm²Vmin, and for 7.5 wt% carbon black suspension in Fig.5, the average deposition efficiency was 0.23 mg/cm²Vmin. Considering that the content of carbon black in suspension is half of the SiC content, there is not so large difference in deposition efficiency between SiC and carbon black powders.

3.3 Laminated composite

SiC/C laminated composite was prepared by alternate deposition from SiC suspension and carbon black suspension. Fig.6 shows the morphologies of the laminated composite before and after hot pressing. As shown, the laminated composite possessed uniform and flat SiC matrix layers separated by carbon interlayers,

demonstrating that typical laminated structure was achieved by EPD method. The thicknesses of matrix layer and interlayer for green compact were about 150 μm and 12 μm, respectively. After sintering, the layer thicknesses of ~75 μm for matrix layer and ~8 μm for interlayer were shown by observation. Larger shrinkage took place in matrix layers than in interlayers, indicating the densification of SiC matrix layers with aid of Al₂O₃ and Y₂O₃ codeposited from SiC suspension. On the other hand, carbon interlayer seemed to be more porous.

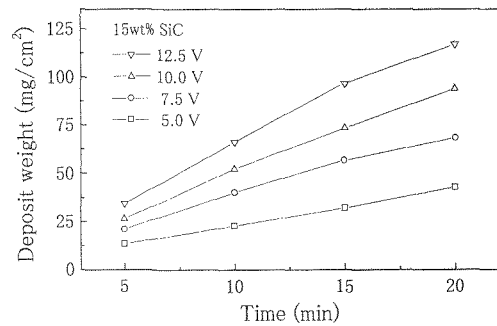


Fig.4 Variation of EPD weight of SiC as a function of time and applied voltage in 15 wt% suspension.

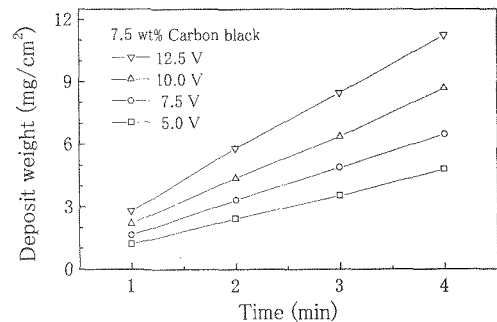


Fig.5 Variation of EPD weight of carbon black as a function of time and applied voltage in 7.5 wt% suspension.

Load-displacement response of the laminated composite in three-point bending test exhibited non-catastrophic fracture process as shown in Fig.7, in which, after reaching the maximum strength, the response curve was zigzagged with some durability. Thus, the carbon interlayer could take a role for deflecting through-thickness crack. The toughness of the composite is expected to be improved by the crack deflection.

4. CONCLUSIONS

A silicon carbide / carbon laminated composite was successfully prepared by electrophoretic deposition technique followed by hot pressing. The comparison of deposition rate of SiC powder between deionized water and acetone medium revealed that the rate in acetone-base suspension was about 18 times larger than that in aqueous suspension. Thus, the acetone medium is more suitable for the deposition of thick SiC layers. The deposit weight of SiC in acetone suspension increased linearly with the increases in applied voltage and

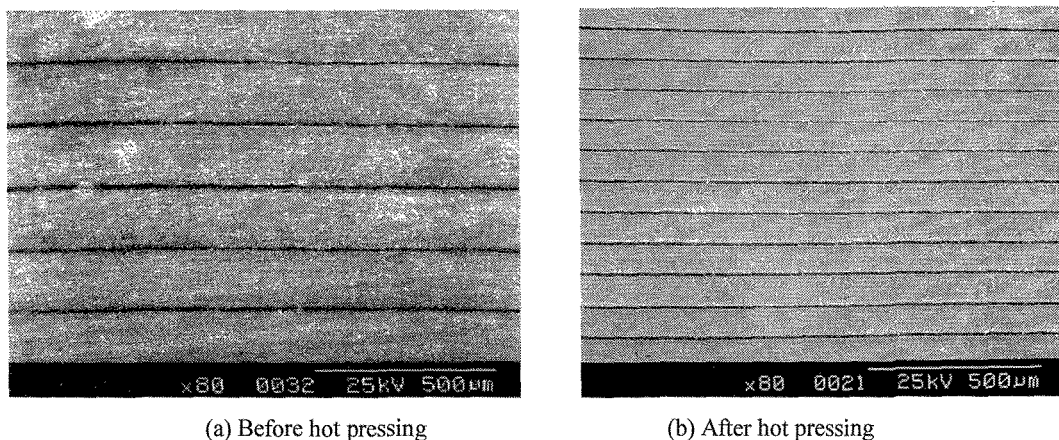


Fig.6 SEM photographs of laminated composite before and after hot pressing.
(Layer number: 48 for SiC and 49 for carbon)

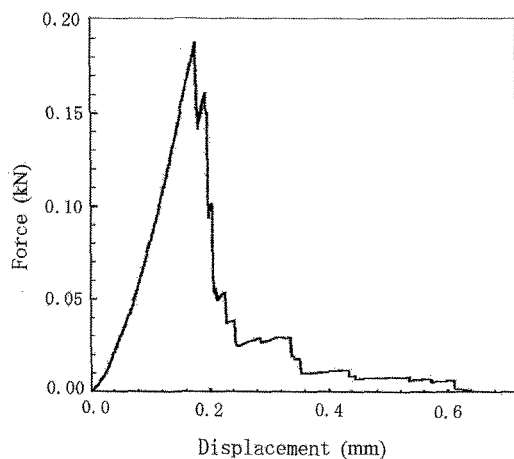


Fig.7 Load-displacement response of the laminated composite in three-point bending test

deposition time. The deposit weight of carbon black powder was also controlled in the similar way. Typical laminated structure with the layer thickness about $75\ \mu\text{m}$ for matrix layer and about $8\ \mu\text{m}$ for interlayer was attained by alternate deposition from SiC suspension and carbon black suspension. Load-displacement response of the laminated composite exhibited non-catastrophic fracture process, demonstrating the possibility for improving the durability of SiC composite by the lamination.

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