ALTERNATING CURRENT ELECTROPHORETIC DEPOSITION (AC-EPD) OF SiC NANOPARTICLES IN AN AQUEOUS SUSPENSION FOR THE FABRICATION OF SiC_f/SiC COMPOSITES

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Nanosized β-SiC (~ 52 nm) particles were successfully infiltrated into the fine voids of a two-dimensionally woven Tyranno^{TM}-SA3 SiC fabric preform using an alternating current electrophoretic deposition (AC-EPD) technique for the fabrication of SiC fiber-reinforced SiC composites (SiC_f/SiC). A well-dispersed suspension containing 20 wt. % SiC nanoparticles was prepared in water after adding 0.5 wt. % dispersant and 0.5 wt. % binder at pH 10. The effect of AC frequency on the microstructure of deposited surface and also the effect of deposition time on the infiltration of a matrix phase into woven SiC-fabrics were examined. The composites fabricated by AC-EPD combined with ultrasound exhibited a high degree of infiltration. These results show that an eco-friend AC-EPD process in aqueous medium can be applied to the fabrication of SiC_f/SiC composites instead of using organic solvents.

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1. Introduction

The fabrication of SiC fiber-reinforced SiC composites (SiC_f/SiC) has attracted considerable attention because of their wide range of high temperature applications, including structural components in gas turbines, heat exchangers, space shuttles, and fusion and advanced fission reactors [1-8]. Different methods have been used to fabricate high-quality SiC_f/SiC composites, such as chemical vapor infiltration (CVI) [8], polymer impregnation and pyrolysis (PIP) [9], nano-infiltrated transient eutectic-phase (NITE) process [10], and reaction sintering (RS) [11]. In particular, CVI and PIP are very slow and costly processes, which also result in an incomplete matrix phase filling into the gaps between fibers. On the other hand, the presence of glassy phases originating from sintering aids and the difficulty in infiltrating matrix particles into the fine voids between fibers are still challenges associated with the NITE process. Therefore, attempts have been made to combine different techniques to enhance the degree of matrix slurry infiltration for the fabrication of dense SiC_f/SiC [12, 13].

The electrophoretic deposition (EPD)-based technique is another suitable method for slurry infiltration into a tightly woven structure [5]. This technique is a fast, simple and efficient infiltration method regardless the shape of the preform, in which charged colloidal particles dispersed in a suitable liquid medium migrate and deposit onto a counter electrode under an DC electric field. A modified process originating from EPD, termed electrophoretic particle infiltration (EPI), has been used to infiltrate ceramic particles into fibrous preforms [14]. Several studies [5–7, 15–17] have used EPD-based techniques to fabricate SiC_f/SiC composites because of the lower manufacturing cost and shorter processing time compared to other methods.

Electrophoretic deposition from an aqueous suspension under an AC electric field (AC-EPD) instead of DC has recently emerged as a successful processing technique for the deposition of a range of particles [18-23], including SiC [24]. In this AC-EPD, the particle velocity shows a

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non-linear dependence under a strong asymmetric AC field, where a particle travels for a longer distance during one of the two half cycles, which results in net particle movement [19, 25]. AC-EPD is advantageous technique for depositing powders with a broad particle size distribution because it can control particle migration according to the wave asymmetry and frequency [18].The effects of the AC frequency on the deposition yield for SiC has been reported elsewhere [24].

The fabrication of SiC_f/SiC composites from aqueous suspensions instead of organic solvents is economically viable from an industrial point of view. Owing to the increasing health and environmental concerns associated with solvent-based systems, research on AC-EPD using aqueous suspensions instead of toxic solvents is needed. Based on previous studies on the optimization of the suspension dispersion and process parameters for the deposition of SiC [24], this paper describes efficient AC-EPD to enhance the degree of slurry infiltration into the fine voids between two-dimensional Tyranno™ fibers for the fabrication of dense SiC_f/SiC composites.

2. Experimental procedure

Two-dimensionally-woven Tyranno™-SA Grade-3 SiC fabrics (Ube Industries LTD., Japan), as reinforcements, were punched into 5 cm diameter discs. Commercial β-SiC (D_m = 52 nm, > 97.5% pure, 4620KE, NanoAmor Inc., USA) was used as the matrix phase. To prepare a stable slurry composed of β-SiC particles for matrix phase infiltration, 0.5 wt. % ammonium polycarboxylate (Cerasperse 5468CF, San Nopco, Korea) and 0.5 wt. % water-soluble acrylic binder (WB4101, Polymer Innovations, USA) were used as the dispersant and binder phase, respectively. After adding the dispersant and binder, the slurry was ball-milled for 24 h using 6 mm SiC balls to ensure particle dispersion. The zeta potential of the SiC suspension was measured using an electroacoustic-type zeta potential analyzer (Zeta Probe, Colloidal Dynamics, USA) after adjusting the pH of the suspension using ammonia and nitric acid.

After preparing a stable aqueous suspension containing 20 wt. % β-SiC nanoparticles, AC-EPD was performed using a dual electrode system for a disc-shaped SiC preform. A programmable function generator (3322A, Agilent, USA), power amplifier (PZD 700A M/S, Trek, USA) and digital oscilloscope (DSO-X 2002A, Agilent, USA) were used for AC signal generation and monitoring. A square-shaped asymmetric waveform with an asymmetry factor of 4, peak-to-peak voltage of 20 V and a 100 Hz frequency were chosen for AC-EPD, based on previous experimental results [24]. Ultrasonic pulses of 10 W with a 1 s cycle were applied using a probe-type ultrasonicator (HD 2070, Bandelin, Germany) to minimize the surface sealing effect. The distance between the SiC fabric and two stainless steel electrodes was 20 mm. The particle size of the starting β-SiC particles and the green microstructure of the matrix deposited on the SiC fabrics after drying at 150°C overnight were characterized by scanning electron microscopy (SEM: S-4200, Hitachi). Transmission electron microscope (TEM: Tecnai G2F20 S-twin, FEI) was also used to observe the particle size of starting nanosized β-SiC particles.

3. Results and discussion

As two-dimensionally woven Tyranno™-SA3 SiC fabrics have large inter-bundle voids and small intra-bundle gaps between each fiber, as shown in Fig. 1(a) and inset of (a), respectively, very fine β-SiC particles would be essential to enhance the degree of matrix phase infiltration. Fig. 1 (b) and inset of (b) presents SEM and its corresponding TEM images respectively, of nanosized β-SiC particles, which were used as the matrix phase in the present investigation. The mean particle size estimated from the SEM and TEM images was approximately 52 nm. Fig. 2 shows the experimental apparatus used for the infiltration of β-SiC particles into the fine voids of the SiC fabric by combined electrophoretic deposition and ultrasonication [17]. One important barrier to be overcome for the fabrication of highly dense SiC_f/SiC associated with matrix phase infiltration is 'surface sealing,' which is a preferential deposit of the matrix phase at the surface without penetrating into the deep voids during EPD. If surface sealing occurs, many unfilled voids will exist in the SiC fabric preform, resulting in a low composite density. Ultrasonic pulses can minimize this surface sealing by releasing the preferentially surface-adsorbed particles and enhancing infiltration into the deep voids of the preform during the EPD process. This study adopted a dual electrode system [17], which was also expected to enhance the degree of infiltration compared to the widely used single electrode system due to deposition from both sides.

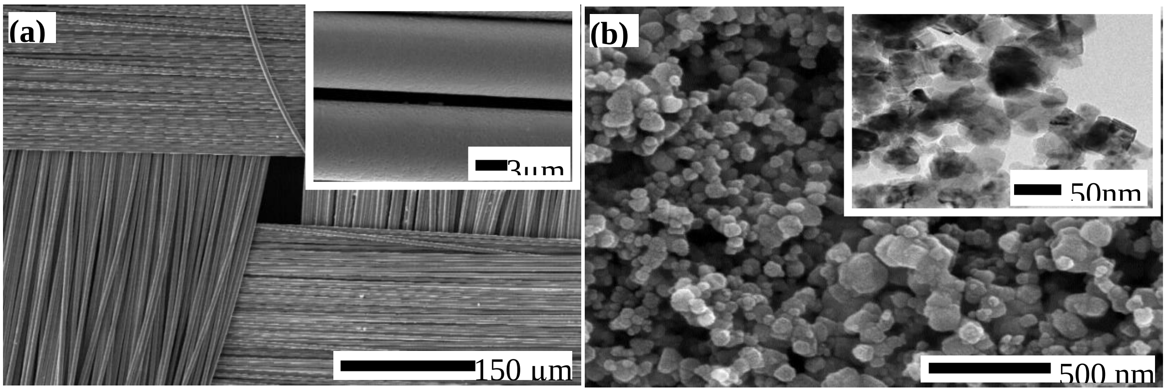

Fig.1. SEM images of Tyranno^{TM}-SA Grade-3 SiC fabrics at two different magnifications ((a) & inset of (a)); (b) SEM image of β-SiC particles (inset shows its corresponding TEM image).

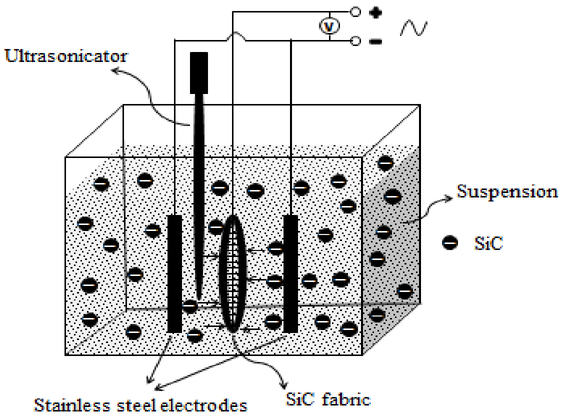

Fig.2. Schematic diagram of the modified experimental apparatus [17] for matrix phase infiltration using AC-EPD combined with ultrasonication.

Efficient EPD begins from the preparation of a stable suspension of homogeneously-dispersed constituent particles. Therefore, the addition of a suitable aqueous dispersant is essential in the colloidal EPD process. The dispersant needs to overcome the strong van der Waals forces between particles and then prevents re-agglomeration. In the present study, ammonium polycarboxylate was used as a dispersant. The surface chemistry and aqueous colloidal stability of the SiC powders with different dispersants including ammonium polycarboxylate is reported elsewhere [5, 26-28]. Moreover, an appropriate amount of binder is also desirable for achieving a stable suspension by steric stabilization due to the adsorbed polymeric layer and for conferring a sufficient adhesion force for deposition. The suspension pH has a significant effect on the zeta potential behavior of SiC particles, showing a decrease in the zeta potential with increasing pH because of the increased hydroxyl ions (OH^-). After adding both of 0.5 wt. % binder and dispersant, the suspension pH was set to 10 for EPD to maximize the surface charge and electrostatic repulsion between particles, whereby the zeta potential for SiC particles was ≤ –55mV [24]. The higher surface charge confers a higher electrophoretic mobility of the particles, which is the principal driving force for EPD under an applied electric field.

The frequency of the AC signal is the main parameter of the deposition yield and surface morphology of the film deposited by AC-EPD. In general, the deposition yield is directly proportional to the applied electric voltage and deposition time, until the depletion of particles begins in the suspension [29]. Therefore, the frequency-dependence on the deposition yield is the most important factor for AC-EPD because different types of particles respond differently to an

AC field. As a preliminary test, EPD was performed on stainless steel electrodes using a suspension containing both 0.5 wt. % binder and dispersant to observe the effects of the AC frequency on the microstructure of deposited SiC films. An examination of the effects of the frequency revealed 100 Hz to be the optimal frequency for achieving the highest deposition rate and green density [24]. Fig. 3 (a) – (f) compares the film surface morphology deposited for 30 minutes using three different typical AC frequencies of 20, 100 and 1000 Hz, respectively, at two different magnifications. According to Fig. 3, the film deposited at 100 Hz showed a smooth surface with a homogeneous particle packing when compared to the films deposited at other frequencies. It can also be clearly seen from the higher magnification microstructural images that the films deposited at 20 and 1000 Hz frequencies are having many SiC particle agglomerations with many pores when compared with the film deposited at 100 Hz frequency. The size distribution of SnO_2 particles deposited under different frequencies by AC-EPD was studied by Raissi et al. [30] and they observed that the deposited particle sizes are inversely proportional to the applied frequency. Therefore, choosing an optimal frequency is very much essential in achieving homogeneous particle deposition in AC-EPD.

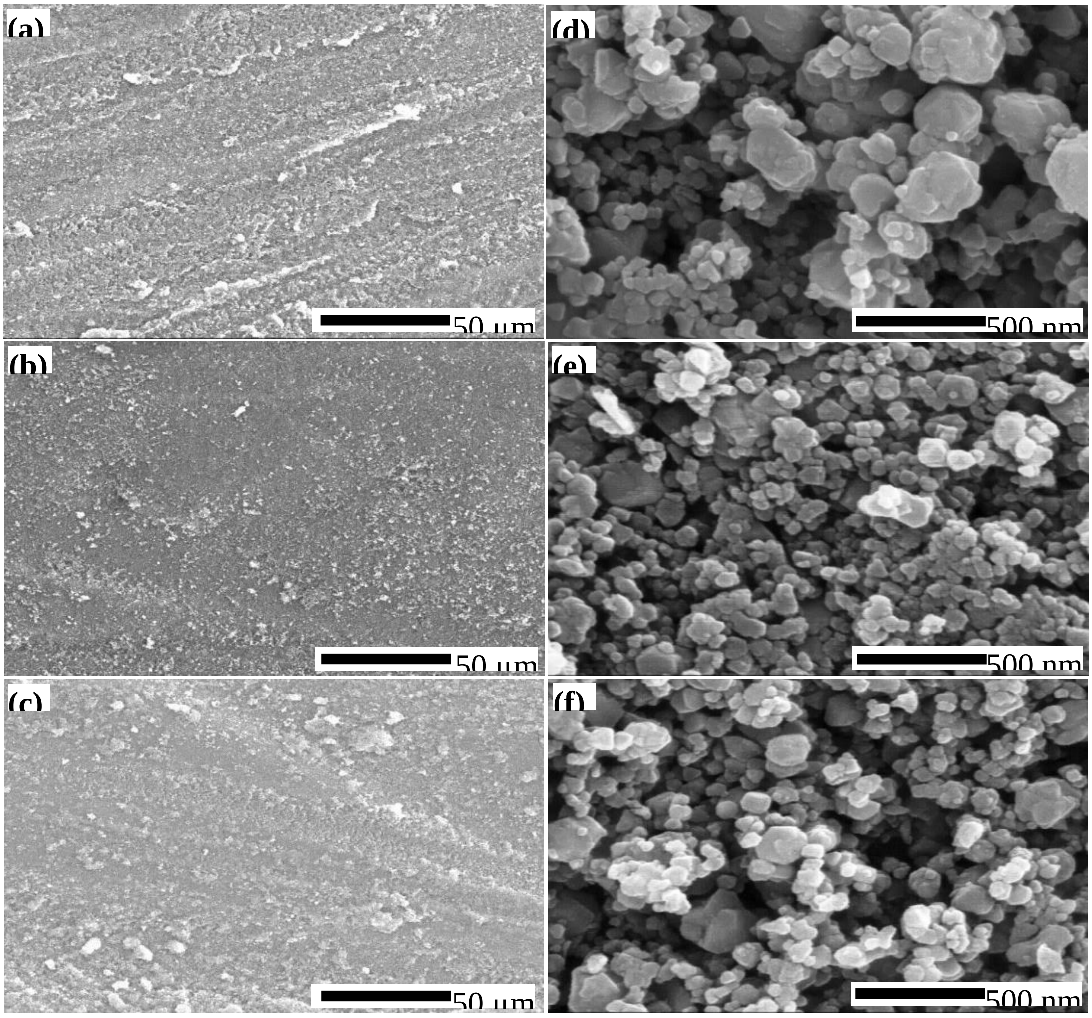

Fig.3. Comparison of the film surface morphology deposited using three different typical AC frequencies at two different magnifications; (a) & (d) 20 Hz, (b) & (e) 100 Hz and (c) & (f) 1000 Hz, respectively.

After examining the effects of the AC frequency, EPD was performed on the SiC-fiber woven fabrics using the same optimal frequency (100 Hz). The effects of the deposition time on the degree of SiC particle infiltration into the woven fabrics was studied. Initially, EPD was performed for 10 minutes by applying ultrasonic pulses for an initial 5 minutes. Fig. 4 (a) shows the corresponding SEM image. Although a certain amount of matrix phase had infiltrated into voids of the woven fabric, the infiltration of particles was not homogeneous and the particles could not attach well to the fibers for this 10 minutes deposition time. On the other hand, when the deposition time was increased to 20 minutes with an ultrasound for the initial 10 minutes, more homogeneous matrix phase infiltration along with the firm attachment of particles to the fabric was observed, as shown in Fig. 4 (b) and inset of (b), at two different magnifications. The matrix phase infiltrated efficiently into the fine voids of the fabrics owing to the vibrating packing mechanism under an AC electric field and the agitation effects of the ultrasonic pulses.

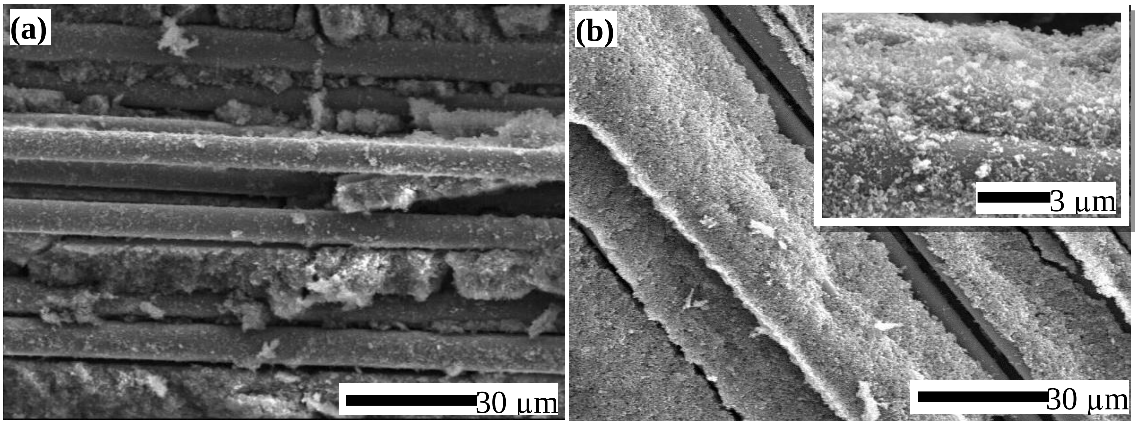

Fig.4. SEM images of a cryo-fractured green fabrics after infiltration for 10 min (a), 20 min at two different magnifications (b) and inset of (b)

To fabricate dense SiC_f/SiC composites with a fiber volume fraction of approximately 50 vol. %, a considerable amount of matrix phase deposition on the surface of fabric is desirable. Therefore, the deposition time was extended to 30 minutes with an initial 20 minutes of periodic ultrasound application, and Fig. 5 (a) and (b) shows the corresponding digital camera and SEM images, respectively. These microstructural observations by SEM (Figs. 4 and 5) clearly suggest that SiC particles were effectively filled the narrow gaps between the individual fibres as well as deposited on the surface of fibres, demonstrates the efficiency of the use of AC-EPD technique to fabricate SiC_f/SiC composites in aqueous suspension. The fabrication of SiC_f/SiC composites with sintering additives by stacking 15 layers of infiltrated SiC fabrics followed by hot pressing is currently underway.

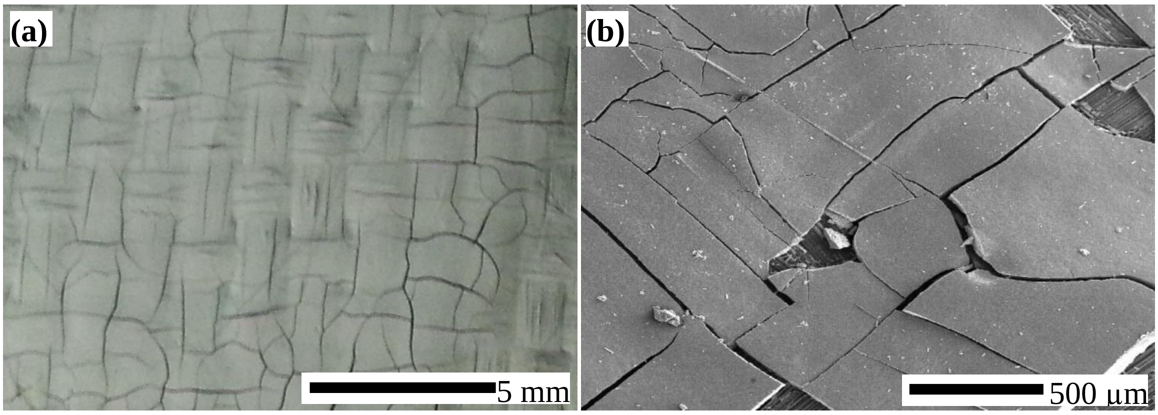

Fig.5. (a) Digital camera image of green fabric after infiltration for 30 min and its corresponding SEM image (b)

4. Conclusions

This paper reported the effectiveness of AC-EPD combined with ultrasonication in an aqueous suspension for the deposition and also infiltration of a nano-sized SiC matrix phase into the fine voids of a two-dimensionally woven Tyranno^{TM}-SA3 SiC preform for the fabrication of SiC_f/SiC composites. Microstructural studies showed that the frequency of AC signal has a pronounced impact on the surface of deposited films and 100 Hz was selected as the optical frequency for the deposition and infiltration of β-SiC nanoparticles. SEM showed that AC-EPD combined with ultrasonication was quite effective in infiltrating the matrix phase containing SiC particles into 2-dimesionally woven SiC fabrics. Overall, eco-friend AC-EPD in aqueous suspensions can be used to fabricate SiC_f/SiC composites instead of organic solvents, which is economically viable from an industrial point of view.

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