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Author(s)	FUJISAWA, Masashi; HATA, Toshimitsu; BRONSVELD, Paul; CASTRO, Vinicius; TANAKA, Fumio; KIKUCHI, Hikari; FURUNO, Takeshi; IMAMURA, Yuji
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Development of SiC/C Composite Materials from Wood Charcoal by a Pulse Current Sintering Method and Their Properties

Masashi FUJISAWA*², Toshimitsu HATA*³, Paul BRONSVELD*⁴,
 Vinicius CASTRO*³, Fumio TANAKA*⁵, Hikari KIKUCHI*^{3,6},
 Takeshi FURUNO*⁷ and Yuji Imamura*²

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Introduction

Wood charcoal has been used for the purification of gas and liquid¹, and adsorption or desorption of moisture, as well as the conventional energy source. Recently it is focused on as a new functional material of thermal characteristics and electrical conductivity², both of which are greatly influenced by the microstructure of carbons during carbonization at high temperatures while pressing, although a great deal of time and energy are required. We have applied a pulse current sintering method to the carbonization of wood charcoal samples at high temperatures as the means of reducing the time and energy required^{3,4}. The electric pulse current can shorten the heating time so that composites can be rapidly produced, and this would make the total production time required very short.

SiC ceramics, which are very strong, and have very high thermal conductivity, heat-resistance, and wearability have been widely applied to industrial materials such as ceramics or alloys. In recent years, a great deal of interest has been generated in research on SiC ceramic composite materials from wood-based biomass⁵⁻¹¹. Some researchers have attempted to develop SiC ceramic composites based on the structure of wood^{12,13}.

In this research, we developed SiC/C composite materials from wood charcoal and SiO₂ powder by a pulse current sintering method. The microstructure, electrical resistivity, and thermal constants of the specimens were examined in order to develop high functional SiC/C composite materials with high electrical and thermal conductivity.

Materials and Methods

A wood sample consisting of Japanese cedar (*Cryptomeria japonica*) which had been ground into 30 mm particles was used. The wood powder was heated at a heating rate of 4°C/min and was reacted at 700°C for 1 h under an Ar gas flow (100 ml/min) in a laboratory-scale electric furnace, and then naturally cooled to room temperature. The wood charcoal powder and SiO₂ powder (Nacalai Tesque Co. Ltd) sized 32–45 μm were prepared by vibration mill and by sieve.

Ten mass%, 30 mass% and 50 mass% of the SiO₂ powder were prepared based on the dry weight of the wood charcoal. After the powder was thoroughly mixed and carefully put into a 10 mm diameter graphite die, it was heated up to 1400°C–1800°C at the rate of 500°C/min, and reacted for 5 or 30 min under an Ar gas flow (0.1 ml/min) using a pulse current sintering apparatus (VCSP-II, Fig. 1). After the reaction, it was naturally cooled to room temperature. Forty MPa of pressure was applied from the beginning of heating, and was released immediately after heating. The temperature was measured at the center surface of the graphite die using an optical

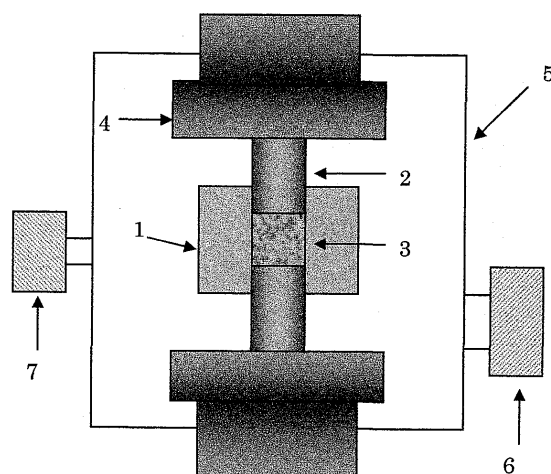


Fig. 1. Schematic diagram of pulse current sintering apparatus. Legend: 1; graphite die, 2; graphite punch, 3; samples, 4; copper plates, 5; chamber, 6; vacuum pump, 7; optical pyrometer.

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*² Laboratory of Deterioration Control.

*³ Laboratory of Wood Composite.

*⁴ Materials Science Centre, University of Groningen, The Netherlands.

*⁵ Laboratory of High Functional Polymers.

*⁶ SS Alloy Co., Ltd.

*⁷ Faculty of Science and Engineering, Shimane University.

pyrometer. The SiC/C was cut into specimens sized $\phi 10$ mm \times approximately 1 mm for each test.

An X-ray diffraction device (Rigaku, RINT-ultra X18) was used to analyze the crystal structure of the specimens under the following measurement conditions; X-ray source: Cu-K α , voltage-current: 30 kV \cdot 100 mA (C-10 mass% SiO₂, C-0 mass% SiO₂), 30 kV \cdot 300 mA (C-30 mass% SiO₂), 45 kV \cdot 300 mA (C-50 mass% SiO₂), respectively.

The composition in the specimens was analyzed by scanning electron microscope (SEM) with the energy dispersive X-ray spectrometer (EDX). The microstructure of the rupture side of the specimens was observed by SEM (JEOL, JSM-5310).

The electric resistivity was measured at room temperature using the four probes method (Loresta HP MCP-T410, Mitsubishi Petrochemical Co. Ltd). The specific heat capacity and the thermal diffusivity of the specimens were measured at room temperature using the laser-flash method, using a thermal-constant analyzer (TC-7000H, Shinku Riko Co. Ltd). Thermal conductivity at room temperature was calculated using the following equation:

$$K = \rho C_p \alpha \quad (1)$$

where K : thermal conductivity ($W \cdot m^{-1} \cdot K^{-1}$), ρ : bulk density ($g \cdot cm^{-3}$), C_p : specific heat capacity ($J \cdot g^{-1} \cdot K^{-1}$), α : thermal diffusivity ($cm^2 \cdot s^{-1}$)

Results and Discussion

The peaks of the β -SiC phase were detected in the X-ray diffraction patterns of the specimens, heated at various heat treatment temperatures with certain SiO₂ contents for 5 and 30 min. The peaks of β -SiC became greater than that of carbon as the SiO₂ contents increased, and this was due to the fact that the amount of active carbon decreased in the formation of β -SiC. It is considered that an optimum content of SiO₂ to carbons is needed to form β -SiC.

It was confirmed by EDX analysis that β -SiC was formed on the surface of the wood charcoal. The shapes of the grains formed were clearly observed in the specimens heated at 1400°C by SEM observation, and pores of various sizes were also observed among the grains. In the specimen heated at 1800°C, the grains were in close contact with each other. The change in structure was caused by the reaction temperature and pressure, which greatly influenced the properties of the products.

The bulk density of the C-10 mass% SiO₂ and C-30 mass% SiO₂ specimens showed a tendency to increase in proportion to the heat treatment temperature for both the 5 min and 30 min reaction time. The bulk density of the C-50 mass% SiO₂ specimens showed different increase patterns for the heat treatment temperature and the

reaction time. It seems that the bulk density increased mainly by the development in the formation of SiC.

The electrical resistivity at room temperature decreased with the increase of the heat treatment temperature in the C-10 mass% SiO₂, C-30 mass% SiO₂, and C-0 mass% SiO₂ specimens, however, the electrical resistivity was independent of the reaction time. The electrical resistivity of the C-0 and C-10 mass% SiO₂ specimens showed similar values. The electrical resistivity of the C-50 mass% SiO₂ specimens showed its maximum in the range of heat treatment temperatures which were affected by the formation of SiC. As the electrical resistance in this experiment shows $10^{-4} \Omega \cdot m$, it is considered that the SiC/C composite is well conducting.

The thermal conductivity at room temperature increased with the increase of the heat treatment temperature in the C-10 mass% SiO₂, C-30 mass% SiO₂, and C-50 mass% SiO₂ specimens, and that of the C-50 mass% SiO₂ specimens showed a greater improvement than that of the other specimens. A maximum of $36 W \cdot m^{-1} \cdot K^{-1}$ was recorded in C-50 mass% SiO₂ specimen at 1800°C for 30 min, which was caused by the formation of the SiC layer on the surface of the wood charcoal grains. As phonon-phonon interaction plays an important role in the thermal conduction of SiC¹⁴⁾, and the layer of SiC was formed in the SiC/C composite materials, the phonon conduction was assumed to be dominant in the specimens.

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