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# Characteristic evaluation of liquid phase-sintered SiC materials by a nondestructive technique

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A B S T R A C T

The nondestructive properties of liquid phase-sintered silicon carbide (LPS-SiC) materials were investigated by an ultrasonic method, in conjunction with the examination of their mechanical properties and microstructures. The damage behaviors of LPS-SiC materials by the cyclic thermal shock were also examined. LPS-SiC materials were fabricated at the temperature of 1820 °C, using the additives of $Al_2O_3$, and $Y_2O_3$ particles. The compositional ratios of additive materials ($Al_2O_3/Y_2O_3$) for LPS-SiC materials were changed from 0.4 to 1.5 with the total amount maintained at 10 wt%. The LPS-SiC materials represented a good density of about 3.2 Mg/m$^3$ and an average flexural strength of about 810 MPa at an additive composition ratio of 1.5. The properties of LPS-SiC materials such as density and flexural strength were more strongly correlated with the attenuation coefficient than with the velocity of ultrasonic wave. The attenuation coefficient of LPS-SiC materials also increased with the increase of thermal shock cycles, reflecting the increased microcrack density.



## 1. Introduction

In the last decades, many efforts were invested to develop the advanced materials for the blanket structures in fusion power plants. SiC fiber reinforced SiC matrix composites ($SiC_f/SiC$) have been extensively studied as a promising material for the blanket first wall and coolant channel [1,2]. A major research for high performance $SiC_f/SiC$ composites has been concentrated on the production of high purity SiC matrix and its densification into the fiber bundle [3–5]. The liquid phase sintering (LPS) process has an advantage for providing a high density of SiC materials, owing to the transformation of additive materials into some eutectics between SiC particles. The mechanical properties of LPS-SiC materials have been improved through the microstructure strengthening by the control of various additive materials [6–8]. The assessment for damage behavior of LPS-SiC materials during the thermal cycles is also necessary for the design of high temperature components. Unfortunately, there are few studies for the thermal shock damages of LPS-SiC materials. It is also important to establish the nondestructive technique by various sources such as acoustic emission, ultrasonic wave and laser probe for the examination of damage degree in structural materials [9–11]. The nondestructive technique has an important role on the investigation of microstructure and mechanical property for LPS-SiC materials. Therefore, the nondestructive data for LPS-SiC materials can be utilized to examine the damage behaviors by the thermal shock and the internal defects by the sintering process.

The purpose of the present study is to characterize LPS-SiC materials fabricated with different composition ratios of additive materials, based on the detailed analysis of ultrasonic properties. The effect of thermal shock cycle on the ultrasonic properties of LPS-SiC materials was also examined.

## 2. Experimental procedures

### 2.1. Fabrication of LPS-SiC materials

LPS-SiC materials were fabricated by a hot pressing process, using a commercial SiC powder (β-type, Ibiden Corp., Japan) with an average size was about 0.3 μm. $Al_2O_3$ and $Y_2O_3$ particles (High Purity Chemical Corp., Japan), whose average sizes were about 1 μm and about 2 μm, respectively, were used as sintering additives. The addition of $Al_2O_3$ and $Y_2O_3$ particles has some advantages for the fabrication of LPS-SiC materials with high density and high strength, even if the elements of Al and Y can reduce the low activation. The complex mixture containing SiC, $Al_2O_3$, $Y_2O_3$ and acetone was prepared by a ball milling process. The blending speed and its holing time for complex mixture were 160 rpm and 12 h, respectively. The total amount of $Al_2O_3$ and $Y_2O_3$ particles were constant as 10 wt%. The compositional ratios ($Al_2O_3/Y_2O_3$) of additive materials were 0.4, 0.7 and 1.5, respectively. LPS-SiC

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materials were fabricated at the temperatures of 1820 °C for creating the eutectoids of $Al_2O_3$ and $Y_2O_3$ particles. The applied pressure and its holding time under the vacuum atmosphere were 10 MPa and 2 h, respectively. The dimensions of as-pressed samples were $3(t) \times 40 \times 40\ mm^3$ and $10(t) \times 40 \times 40\ mm^3$, respectively.

### 2.2. Characterization evaluation of LPS-SiC materials

The sintered densities of LPS-SiC materials were determined by the immersion method, according to Archimedes' principle. The microstructure of LPS-SiC materials was also analyzed using a scanning electron microscope (SEM). The three point bending test for LPS-SiC materials was carried out at room temperature. The dimension of test samples for three point bending test was $3(t) \times 4 \times 25\ mm^3$. The span length and the crosshead speed were 18 mm and 0.5 mm/min, respectively. The thermal shock test for LPS-SiC materials was carried out at the temperature of 250 °C. The test sample was heated to operating temperature for 20 min, prior to the dropping into the water. The thermal shock test was repeatedly performed up to 15 cycles.

The nondestructive technique by an ultrasonic wave was induced to investigate the properties of LPS-SiC materials. The damage behavior of LPS-SiC materials suffered from the cyclic thermal shock was also examined by a nondestructive test. An ultrasonic wave propagating through a solid body can be used to measure the properties of materials. Both the velocity and the attenuation coefficient of ultrasonic wave are changed by the homogeneity of materials, since these results from the variation of acoustic impedance and wave energy. The velocity of ultrasonic wave was determined by the thickness of test sample and the time between front echo and backwall echo in the waveform. The attenuation coefficient of ultrasonic wave was also calculated from the magnitude of front and backwall echoes. The ultrasonic properties for LPS-SiC materials were measured by the water-immersion method. The nondestructive test system is composed of pulse/receiver device, oscilloscope and computer for the exact measurement and the analysis of ultrasonic wave. The frequency range of transmitter was constant as 10 MHz. The dimension of test sample was $10(t) \times 20 \times 25\ mm^3$.

## 3. Results and discussion

Fig. 1 shows the compo-micrographs of LPS-SiC materials fabricated with the additive composition ratios of 0.4 and 1.5. LPS-SiC materials represented a good microstructure without sintering defects like microcracking. The morphology of LPS-SiC materials was mainly composed of SiC phases of gray area and secondary phase of white area, accompanying some amount of micropores. The inhomogeneous dispersion of secondary phases around SiC phases was observed at the additive composition ratio of 0.4. The amount of micropores in the microstructure also tended to decrease with the increase of additive composition ratio.

The density and the flexural strength of LPS-SiC materials by the variation of additive composition ratios were summarized in Table 1. The relative density of LPS-SiC materials was determined as a ratio of measured density to the theoretical density, which was calculated by the rule of mixture. The relative density of LPS-SiC materials tended to increase with increasing the compositional ratio of additive materials. Especially, LPS-SiC materials had an excellent relative density of about 98% (about 3.2 $Mg/m^3$) at the additive composition ratio of 1.5. This is maybe because the amount increase of $Al_2O_3$ particles with a melting point lower than that of $Y_2O_3$ activates the creation of secondary phases such as YAG and $Al_2O_3$-YAG around SiC particles. [7] Moreover, the increase of additive composition ratio for LPS-SiC materials led to the increase of flexural strength. LPS-SiC materials had an average flexural strength of about 810 MPa at the additive composition of 1.5, accompanying the increase of sintered density associated with the formation of secondary phase.

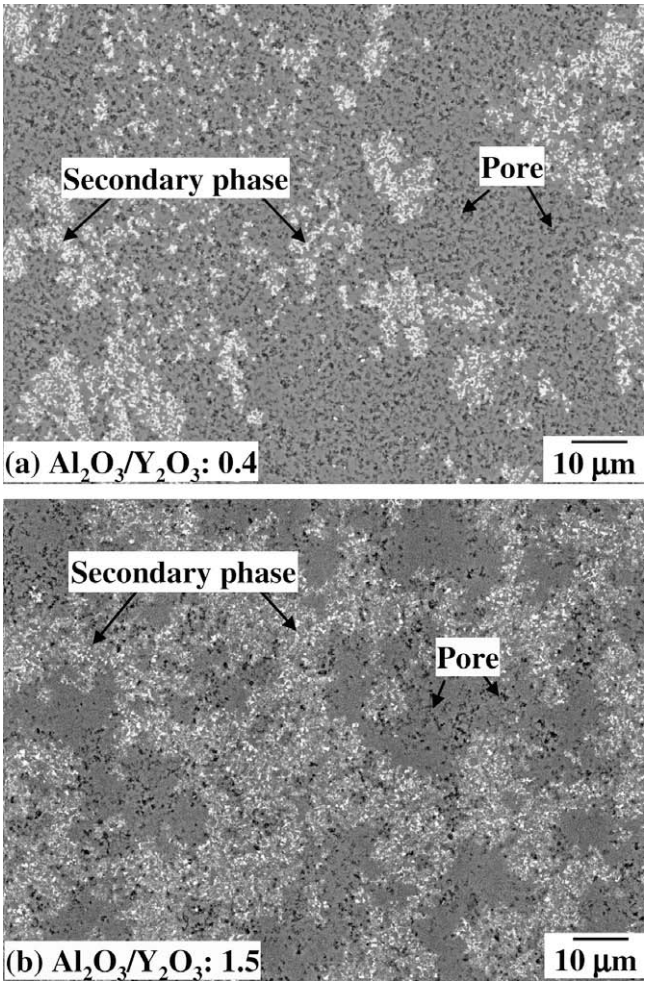


**Fig. 1.** Microstructures of LPS-SiC materials fabricated with different composition ratios of additive materials.

**Table 1**
Characterization of LPS-SiC materials by the variation of additive composition ratios.

| Additive composition ratio ($Al_2O_3/Y_2O_3$) | Measured density ($g/cm^3$) | Flexural density (%) | Flexural strength (MPa) |
|---|---|---|---|
| 0.4 | 3.1 | 93.4 | 420 |
| 0.7 | 3.2 | 95.8 | 780 |
| 1.5 | 3.2 | 97.9 | 810 |

Fig. 2 shows the effect of additive composition ratio on the velocity of ultrasonic wave and its attenuation coefficient for LPS-SiC materials. The ultrasonic wave properties of LPS-SiC materials depended on the compositional ratio of additive materials. The ultrasonic wave velocity of LPS-SiC materials tended to increase with increasing the compositional ratio of additive materials. On the contrary, the increase of additive composition ratio for LPS-SiC materials led to a great reduction of attenuation coefficient. LPS-SiC materials represented an ultrasonic wave velocity of about 9650 m/s and an attenuation coefficient of about 35 dB/m at the additive composition ratio of 1.5. Generally, the attenuation coefficient of LPS-SiC materials was related with the densitification of microstructure by the variation of additive composition ratios.

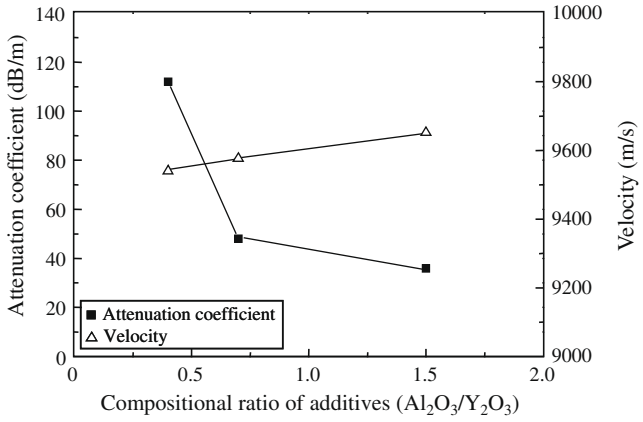


**Fig. 2.** Effect of additive composition ratio on the velocity of ultrasonic wave and its attenuation coefficient for LPS-SiC materials.

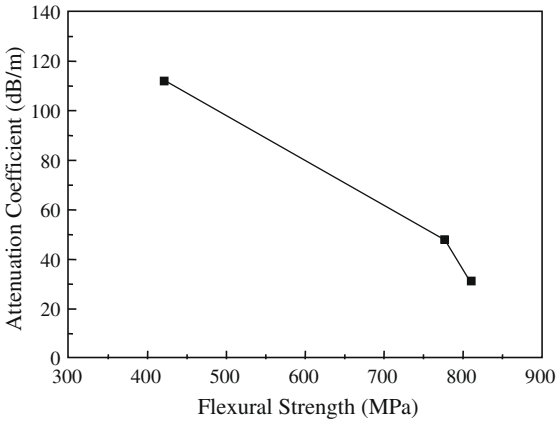


**Fig. 4.** Relationship between flexural strength and attenuation coefficient in LPS-SiC materials.

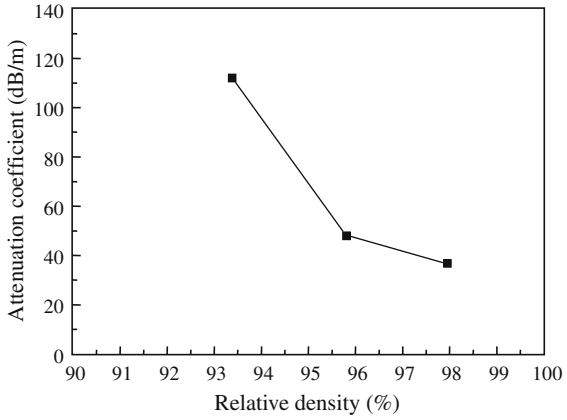


**Fig. 3.** Relationship between relative density and attenuation coefficient in LPS-SiC materials.

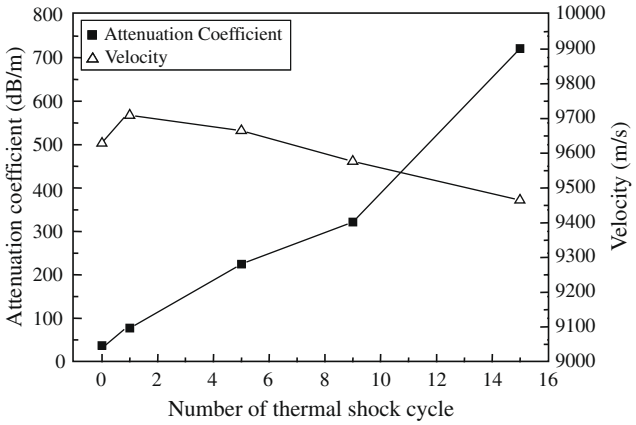


**Fig. 5.** Effect of thermal shock cycle on the velocity of ultrasonic wave and its attenuation coefficient for LPS-SiC materials.

Fig. 3 show the relationship between relative density and attenuation coefficient of LPS-SiC materials fabricated with different composition ratios of additive materials. The relative density of LPS-SiC materials was shown in Table 1. The attenuation coefficient of ultrasonic wave for LPS-SiC materials decreased with the increase of relative density. Especially, LPS-SiC materials represented an average attenuation coefficient of about 105 dB/m at the relative density of about 93%, corresponding to 3 times of that at the relative density of about 98%. This is maybe because the suppression of sintering defects like void or pore in the microstructure reduces the scattering of ultrasonic wave.

Fig. 4 show the relationship between flexural strength and attenuation coefficient of LPS-SiC materials fabricated with different composition ratios of additive materials. The flexural strength of LPS-SiC materials strongly depended on the attenuation coefficient of ultrasonic wave, even if there are limited data in the experiment. In other words, the flexural strength of LPS-SiC materials linearly increased with the decrease of attenuation coefficient for the reflected ultrasonic wave. It seems that the nondestructive technique for the sintered body can be regarded as an another effective method for predicting the mechanical properties of LPS-SiC materials.

Fig. 5 show the effect of thermal shock cycle on the velocity of ultrasonic wave and its attenuation coefficient for LPS-SiC materials. LPS-SiC materials were fabricated with the additive composition ratio of 1.5. It was found that the attenuation coefficient of ultrasonic wave was very sensitive for the thermal shock damages

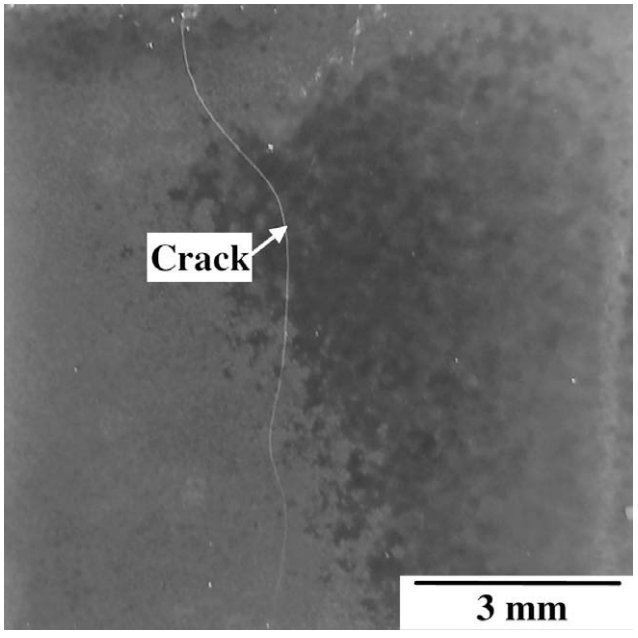


**Fig. 6.** Macrostructure of LPS-SiC materials by the thermal shock cycle of 15 numbers.

of LPS-SiC materials. LPS-SiC materials maintained the level of ultrasonic wave velocity form about 9450 m/s to about 9700 m/s. On the other hand, the attenuation coefficient of ultrasonic wave for LPS-SiC materials was greatly affected by the variation of thermal shock cycles. The increase of thermal shock cycles led to a rapid increase of attenuation coefficient for LPS-SiC materials. Especially, LPS-SiC materials represented an attenuation coefficient of about 700 dB/m after the thermal shock of 15 cycles, which corresponded to about 20 times of as-pressed materials without a thermal shock. This is maybe related with the damage of microstructure, as shown in Fig. 6. In other words, such a variation of nondestructive properties is due to the creation of microcrack and its extensive propagation by the repetitive thermal shock.

## 4. Conclusions

The relative density of LPS-SiC materials increased with the increase of additive composition ratio, accompanying the decrease of micropores and the uniform dispersion of secondary phases by the chemical reaction of $Al_2O_3$ and $Y_2O_3$ particles. Especially, LPS-SiC materials represented an relative density of about 98% and an flexural strength of about 810 MPa at the additive composition ratio of 1.5.

The increase of additive composition ratio for the fabrication of LPS-SiC materials resulted in the increase of ultrasonic wave velocity and the reduction of ultrasonic wave attenuation coefficient. Especially, the characterization of LPS-SiC materials was greatly affected by the attenuation coefficient of ultrasonic wave. The improvement of relative density and flexural strength for LPS-SiC materials led to the decrease of attenuation coefficient, owing to the reduction of ultrasonic wave scattering. The attenuation coefficient of LPS-SiC materials rapidly increased with the increase of thermal shock cycles, due to the creation of microcrack and its propagation. Therefore, it is considered that the thermal shock damages and the sintering properties of LPS-SiC materials can be nondestructively investigated by evaluating the attenuation coefficient of ultrasonic wave.

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