Fabrication of porous SiC mat for radiation heater by controlling rheology of polycarbosilane

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Introduction

Combustion properties such as flame stability and flame structure include CO and NO_x emissions in a gas-radiation heater are affected by controlling the optical thickness, scattering properties, fuel gas permeability and thermal conductivity of the porous radiant plate because combustion happens inside and surface of a radiant plate in the form of free flame and combustion energy is transferred to the plate making an infra-red radiation. Fibrous radiant plate have been developed to make efficient radiant systems for uniform heating of object surface with complete and clean combustion and the promising one is porous SiC fiber mat which having a high specific surface area and a high radiation efficiency due to its black colored body

In this study, we have investigated an efficient way of forming porous SiC fiber mat from a simple in-situ process using a melt blown process and subsequent bonding of the melt blown fibers together by the curing and pyrolysis under the minimum amount of applied pressure. The thermal and mechanical properties of SiC mat were investigated related to the rheology and the radiation effect was also investigated for the application in a gas fired radiation burner.

Experimental

PCS was synthesized from the kumada rearrangement of polydimethylsilane (PDMS) under normal pressure in the presence of wired or powdery solid acid catalyst.17-19 The mixture of 1 kg of PDMS and 10 g of catalyst was loaded into the reaction vessel. After purging the sample with argon gas, the reaction vessel was heated to 350°C and 400°C in two-step for the conversion of PDMS into PCS and subsequent polymerization. It was further polymerized at 400°C for 5-10 h without catalyst after distilled out of the low molecular portion to get the proper rheology for melt blown process. The rheology of PCS was characterized by measurement of melting point using spindle type apparatus, molecular weight distribution (GPC) and melt viscosity.

When the system was reached to spinning condition, the PCS melt was spun out of the nozzle through the gear pump and the hot-air hits the spun melt into winding receiver (metal mesh). Spun mats were subjected to the thermal curing at 200°C for 1 h in a muffle furnace with uniform pressing at low pressure for inducing the point contacts among fibers and then sintered at a temperature of 1200°C for 1 h under an Ar atmosphere in a graphite furnace. The microstructure and morphology of the melt blown mat and that of a single fiber were observed using FE-SEM operating at 15 kV. TGA was used to analyze the pyrolysis behavior of the SiC mat. The gas radiation heating test was performed by the measurement of the temperature profile of the SiC mat covered with the glass plate and the exhaust gas was measured using GC.

Results and Discussion

Figure 1 shows optical images of (a) the precursor mat and (b) SiC mat pyrolyzed (a) at 1200°C. The precursor mat has transparent yellow color and was cut into circular shape with diameter of 200 mm before curing. It pressed down into 2.0 kg/cm^2 during the curing process and pyrolysis at 1200°C under inert atmosphere spontaneously. It reduced to 150 mm after pyrolysis. In Figure 2, fibers in the mat have the circular cross-section and were shown randomly distributed. Every layer was uniformly stacked through the transverse directions like a fagot and each layer and also connected through the point contacts. These contacts, so-called "the neck" at the contact points, were formed in the crossing or parallel patterns and distributed uniformly all over the SiC mat.

SiC mat was flexible under sufficient load as a result of 3-point bending test and this is due to the effect of the "point contact structure" on the relative strength and the flexibility for each mat.

(a) Precursor mat **(b) SiC mat**

Fig. 1 Photograph images of (a) preceramic fiber mat (after curing in air) and (b) SiC mat pyrolyzed (a) at 1200°C.

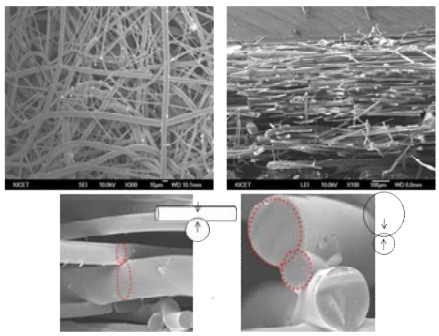

Fig. 2 FE-SEM images of SiC mat pyrolyzed the preceramic fiber mat pyrolyzed at 1200°C for 1 hour

Figure 3 shows the XRD pattern of SiC mat derived from precursor polymer pyrolyzed at 1200°C after cured at 200°C for 1 h under mild compression. The diffraction peaks at around 35.7°, 60°, and 71.8° corresponded to cubic β-SiC which were attributed to the (111), (220), and (311) planes. All the peaks were quite broad because the nanocrystalline β-SiC are dispersed among amorphous Si-O-C matrix which indicated that the SiC fiber was in the early stage of crystallization at this heat treatment temperature. The second phase such as graphite or silica couldn't be found in the XRD pattern.

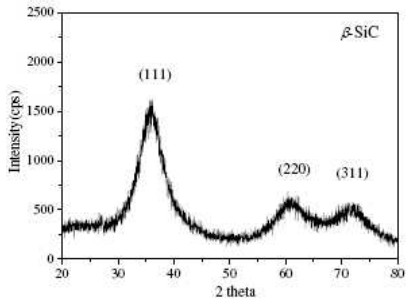

Fig. 3 X-ray diffraction pattern of SiC mat pyrolyzed the PCS mat derived from preceramic fiber mat at 1200°C for 1 hour

SiC mat was examined the oxidation behavior maintained in the atmospheric muffle furnace for 1-48 h at 1000°C. There was no significant difference in weight change during the oxidation until 48 h. The fracture surface of the fiber oxidized at 1000°C for 48 h were shown uniformly coating layer of about 380 nm and the thickness was constant along the fiber periphery without any cracks or bubbles at the boundaries. The oxide layers were also observed at the neck and there wasn't any degradation even at the sharp node. It is expected that uniform layer will slow down.

The infra-red radiation effect of the SiC mat was tested using commercial gas burner. SiC mat was equipped on top of the combustor covered with the flame protecting glass and operated at the adjusted gas mixing ratio (butane : air). Infra-red was radiated all over the mat during the operation. The temperature was also measured by the IR spectrum and radical analysis through gas combustion tester as shown in the Figure 4. and the temperature on the glass reached to 870°C. It is sufficiently heated up to high temperature which was required to burn a fuel gas completely. The results of GC analysis support this complete combustion behavior in the mat. Figure 13 shows the combustion gas evolution with cross-sectional distance of the SiC mat. Carbon monoxide gas was detected to 0 ppm where oxygen gas was reached to 2 percent. If insufficient oxygen supply into the SiC mat makes carbon monoxide gas in the luminous (flame orange color) region. Howeverv carbon monoxide gas is not evolved because of the stable radiation of flame through the optimum oxygen supply condition and it makes the sufficient high surface temperature of the radiation mat. It can be considered that the pore size in the mat was well controlled for fuel gas to flow easily

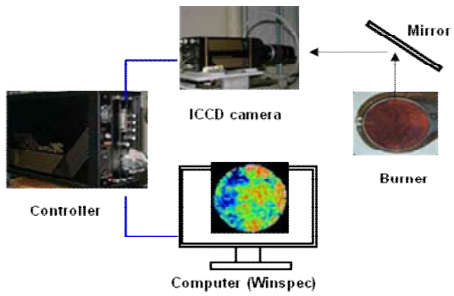

Fig. 4 Experimental setup for IR spectrum and radical analysis

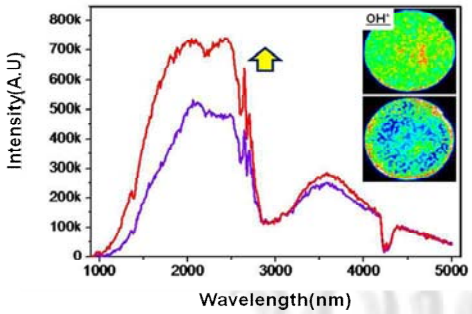

Fig. 5 IR spectrum and radical analysis of SiC fiber mat during gas radiation test

Conclusions

A porous SiC mat was successfully fabricated by melt blowing and pyrolysis of a rheology controlled preceramic polymer. The SiC mat had a bimodal fiber distribution from 1-3 μm to 7-15 μm. Each layer was stacked well through the transverse directions and chemically bonded at the contact points. This bond structure was affected by the rheology of PCS and made a SiC mat very flexible and strong enough for handling during the operation. Furthermore, oxidation layer was formed uniformly on the fiber periphery without any cracks or pores at the boundaries and it will act as an excellent thermal protective layer. Finall IR radiation was observed all over the mat surface. However surface temperature was lower than conventional one and CO gas was evolved due to the incomplete combustion during the burning process.

Acknowledgment.

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References

[1] Tong, T.-W. J Quant Spectrosc Radiat Transfer 1995, 53, 235

[2] Riu, D.-H.; Shin, D.-G.; Kong, E.-B.; Kim, Y.; Hong, H.-J.; Cho, K.-Y.; Huh, S.- H. Ceramist, 2007, 4, 54

[3] Kulkarni, M.-R.; Peck, R.-E. Proc of Joint Meeting of Central and Western States Section and Mexican National Section of the Combustion Institute and American flame Research Committee, 1995, 26.