FABRICATION OF CARBON/CARBON COMPOSITES BY PRESS CARBONIZATION

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INTRODUCTION

A high yield of matrix precursor should be obtained during carbonization to make a high-density carbon/carbon(C/C) composite and therefore to shorten its densification process afterward. In case of pitch as matrix precursor, pressurized carbonization using autoclave [1,2] or HIP [3], modification of pitch by polymerization [4], and addition of high-yield carbon powders [5] have been proposed.
In this work, a new approach to obtain high-density C/C composites was studied theoretically and was verified experimentally.

THEORETICAL CONSIDERATION

Solid pitch, upon continuous heating, melts through a glass transition point, forms mesophase and finally solidifies again into coke. The average molecular weight of the pitch increases during the process by volatile removal, pyrolysis, polymerization and condensation of the pitch molecules. This thermal behavior of pitch may suggest that C/C composites of higher density could be fabricated by press carbonization, in which some volatiles in the pitch matrix of a green composite are first removed by heating, then the green composite is pressed uniaxially to fill the pores formed during the heat-treatment, and finally the heat-treated composite is carbonized still under the press.
Fig. 1 shows schematically the advantage of the press carbonization method over the simple carbonization method.

EXPERIMENTAL

A prepreg was prepared from 8 harness/satin weave carbon cloth impregnated with coal tar pitch. Green composites, 15 to 30mm thick, were then fabricated by stacking and hot-pressing the prepregs. The green composites were heat-treated at 480°C for varying times, and then pressed to 15mm thickness, followed by two step carbonization at 600°C and at 1000°C for an hour, respectively.

RESULTS AND DISCUSSION

Fig. 2 shows the densities of C/C composites fabricated by press carbonization plotted against pressing ratio. The trend in variation of the experimental density corresponds with that of the theoretical one calculated, which increases with an increasing pressing ratio upto 1.5g/cm^3 and then becomes constant.
Slightly lower values of experimental densities than are expected in the theory are due to insufficient heat-treatement before pressing.
In order to get an optimum heat-treatment condition, two kinds of routes are tried, a constant heating-rate and a constant temperature.

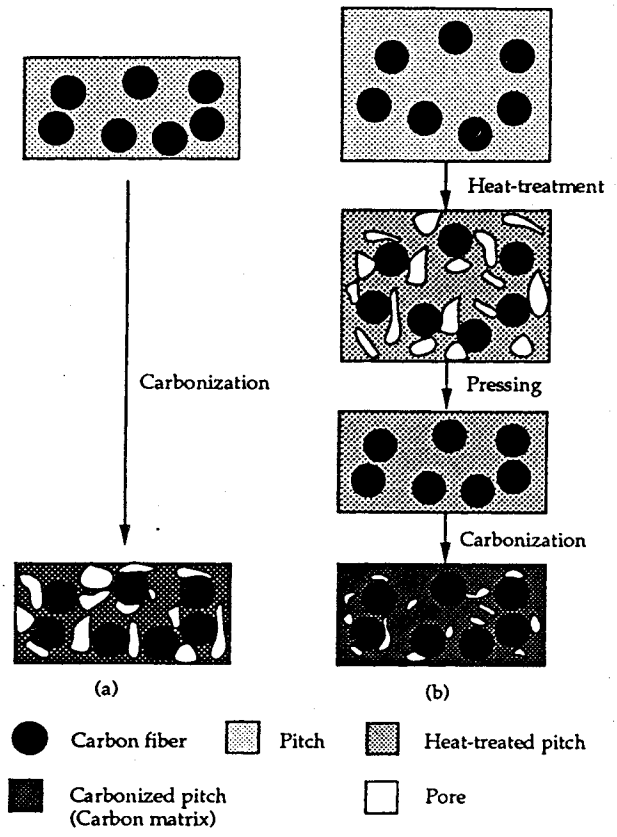

Fig. 1 Schematic diagram of a simple carbonization (a) and press carbonization(b).

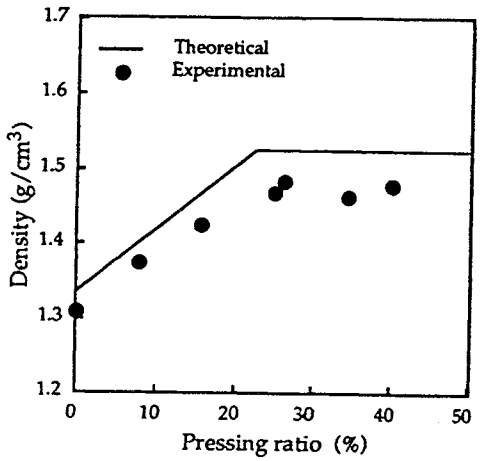

Fig. 2 Theoretical and experimental densities of C/C composites as a function of pressing ratio.

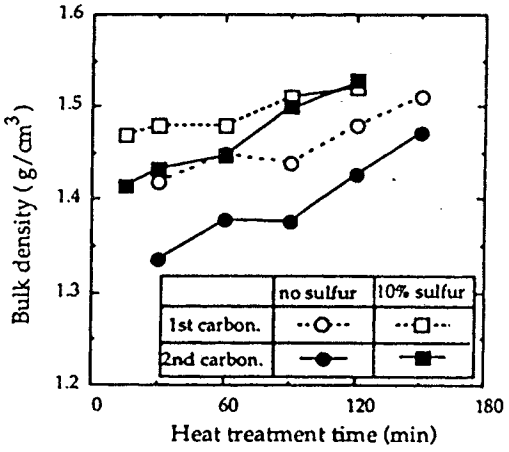

Fig. 4 Density of C/C composites as a function of heat treatment time.

Fig. 3 shows density variation of C/C composites pressed at various temperatures with a heating rate of 5°C/min. The highest density is shown around 630°C, above which the matrix pitch may start to solidifiy.

Fig. 4 shows density variations of C/C composites with pitch or sulfur-added pitch as matrix precursor, which were pressed just after heat-treatment at 480°C for various durations and then carbonized twice at 600°C and 1000°C. The densities increase with increasing heat-treatment time. The composites with sulfur-added pitch show higher densities at the same durations than those with only pitch, resulting from the higher carbonization yield of the matrix. Moreover, while the density shows a large decrease during the 2nd carbonization due to an expansion of the composite with pitch, the density decrease could be diminished by a shrinkage of the composite with sulfur-added pitch.

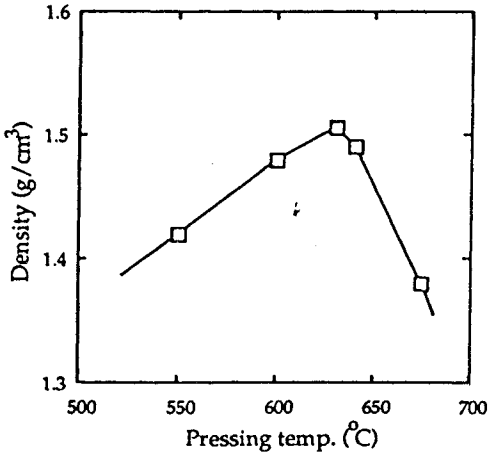

Fig. 3 Density of C/C composites as a function of pressing ratio.

CONCLUSION

The press carbonization method proposed in this study, which consists of heat treatment, uniaxial pressing, and carbonization process, was effective to preparing high-density C/C composites, utilizing the thermal characteristics of pitch matrix.

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