EFFECTS OF PRESSING CONDITIONS ON SINTERING OF CARBON

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Introduction

This paper presents a study of a potential low-cost processing route to carbon-carbon composites [1-2]. Specifically, we examined the compaction and sintering behavior of mesophase pitch powders.

Experimental

AR mesophase pellets (synthetic naphthalene-based mesophase pitch, Mitsubishi AR) as received were jet-milled with air using No. 4 Micro-Jet from Fluid Energy Aljet, spread into uniform beds less than 10 mm deep, then heated to 220°C at 1.5°C/min and held for varying lengths of time to achieve the desired weight gain. Oxidized beds were crushed and mixed, then jet-milled again and compacted according to Table 1. A randomized multivariate statistical design was employed to empirically determine relative effects of important process variables. A 2x6 in (51x152 mm) mold was filled with 25g of evenly distributed powder, closed and optionally heated before applying pressure for various lengths of time as listed in Table 1.

Column		1	2	3	4	5
	Time	Temperature (°C)				
Row	(min)	25	75	150	225	300
A	15	10000 6	1000 2	5000 8	15000 10	300 4
B	30	300 8	15000 6	1000 10	10000 4	5000 2
C	60	5000 10	10000 8	15000 4	300 2	1000 6
D	120	15000 2	5000 4	300 6	1000 8	10000 10
E	300	1000 4	300 10	10000 2	5000 6	15000 8

Table 1. Hyper-greco-latin square matrix of testing conditions showing pressure (psi) and oxidation weight gain (%) for each time (rows) and temperature (columns). (1000 psi = 6.9 MPa)

Compacts were heated in a carbonization furnace at 1°C/min. to 850°C in a nitrogen atmosphere at ambient pressure. A JEOL 840 SEM was used to characterize differences in the compacted and sintered microstructures. FTIR chemical analysis of oxidized powders was performed using 2% pitch in KBr. Relative thermoplasticity measurements (similar to Hoffman's test [3]) were made using a modified TMA apparatus with 60° conical tip probe and 2 g load. Bulk density of the products was determined by Archimedes displacement. Flexural strength was determined using four-point flex test with the maximum l/d ratio possible (16:1 to 32:1). Flexural modulus was determined by using a strain gage on the tensile surface.

Results and Discussion

Warm pressing was shown to be successful in the production of sinterable compacts of oxidized mesophase powder. Although compacts produced with 2% and 4% weight gain bloated, all compacts produced with 6% or higher weight gain did not bloat during the sintering step.

Comparison of thermoplasticity data with literature values [3] was of limited value due to the 400°C upper temperature limit of the TMA apparatus and the higher heating rate (10°C/min) used.

The full temperature range was found to be of interest, rather than just the penetration portion, due to slight changes in Tg and a nonlinear CTE. Bloating was observed as rapid expansion and its onset could be detected prior to penetration. Upward changes in the slope of the thermal expansion curve, such as is seen in the curve for 6.2% weight gain in Figure 1, could be due to the formation of internal porosity as a prelude to bloating.

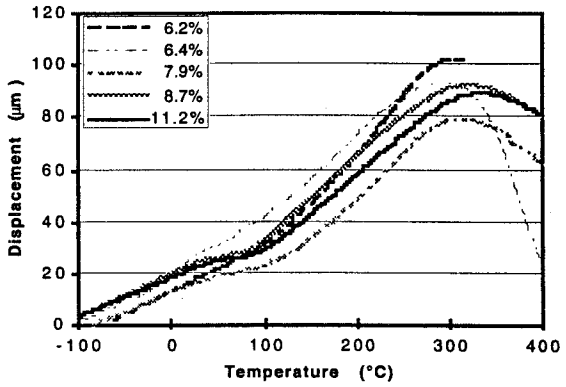
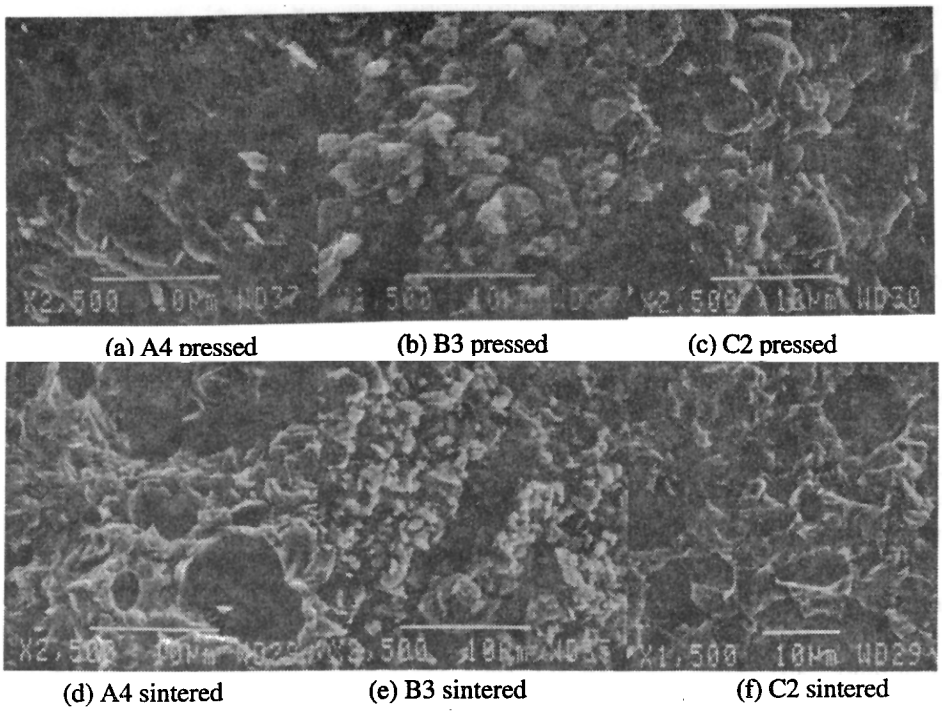


Figure 1. Thermoplasticity changes as a function of isothermal oxidation weight gain in air at 220°C.

FTIR studies included comparison of peak area ratios of carbonyl and ether bond absorptions to the assumed constant aromatic stretching band absorption [4]. Changes were notable after initial oxidation and when mixing unoxidized with oxidized pitch, but



(a) A4 pressed (b) B3 pressed (c) C2 pressed

(d) A4 sintered (e) B3 sintered (f) C2 sintered

Figure 2. Microstructure before and after sintering for conditions shown in Table 1

distinct differences between two and ten percent weight gain were not quantifiable by this method.

Compacts in which local viscous flow occurred at particle contacts were identified by SEM. A partial consolidation is desirable, but particle morphology and open porosity should be maintained. Figure 2 shows typical compacted and sintered microstructures for: A4 (a. and d.) over consolidated, B3 (b. and e.) under compacted, and C2 (c. and f.) adequate compaction.

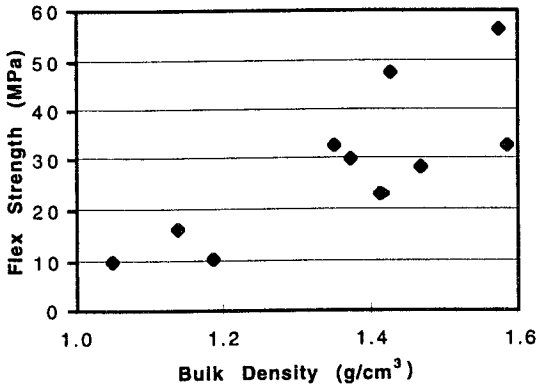


Figure 3. Flexural strength vs. bulk density for sintered carbon.

Due to it's inherent limitations and inability to define additional parameters, the multivariate statistical analysis proved inconclusive for this initial set of test conditions. Effects of changing process conditions were not directly correlated with properties. This is likely due to the scale of the interaction being larger than the range where the multivariate analysis is most sensitive to interactions. Figure 3 shows that oxidation and pressing conditions have some effect on strength independent of final density.

Conclusions

Warm pressing was shown to be successful in the production of sinterable compacts of oxidized mesophase powder. Six percent or higher weight gain was required to prevent bloating. The hypothesis that powders with higher oxygen content require higher temperatures and pressures for compaction and have decreased ability to flow during sintering was apparent, even though it was not proven statistically. The wider processing range which warm pressing allows should enable improved conditions for the incorporation of fiber reinforcement, including higher char-yield and lower shrinkage mesophase powders. The fine scale of the microstructure may also enable controlled matrix cracking eliminating the large intrabundle cracks common to carbon-carbon composites and greatly reducing the grain size of the anisotropic pitch matrix.

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