PREPARATION OF MESOPHASE PITCH-BASED HIGH THERMAL CONDUCTIVITY CARBON MATERIALS

Weidong CHI, Yinying ZHANG, Zengmin SHEN and Hui LIU

Key Laboratory of carbon fiber and functional polymers,
Ministry of Education
Beijing University of Chemical Technology
Beijing 100029, China

Introduction

With the rapid development of technology, more and more electronic devices become smaller, lightweight, compact structure and running highly efficient [1], leading to produce and accumulate large amounts of heat during running process. Therefore, light weight, high thermal conductivity materials are needed urgently. Carbon materials with high thermal conductivity, low density, and low thermal expansion coefficient properties become a promising thermal management material in recent years [2-4]. Mesophase pitch-based carbon material has excellent thermal and electrical conductivity due to its high degree of preferred orientation and less lattice defects [5-8]. As we all know, catalytic graphitization can greatly improve the graphitization degree, which can increase the thermal conductivity of graphite materials. In this paper, catalytic graphitization of mesophase pitch for high thermal conductivity materials was studied.

Experimental

Mesophase pitch was spun into pitch fiber, and then oxidized at different temperature. Put the oxidized pitch fiber into a steel die, and molding at 25MPa, 400°C for 1 hour to form a self-sintered plate. The plate was carbonized at 1000°C, under protection of nitrogen to obtain carbon plate. Catalyst was introduced by immersion the carbon plates into different catalyst system. For example, immersed the carbon plate in 1.33mol LH_3BO_3 solution, and soaking 24h could introduce boron catalyst. We use $TiCl_4$: C_2H_5OH = 0.228:17.2 solution, with soaking 30h to obtain Ti catalyst system, and used $TiCl_4$: H_3BO_3: C_2H_5OH = 0.228:1.33:17.2 solution, soaking 30h to obtain Ti-B composite catalyst system. The soaked plated was dried at 100°C 2 hours. Finally, the high thermal conductivity carbon plate was manufactured by heat treat the carbon plate under the protection of argon gas at 2600ºC.

Results and Discussion

The oxidization temperature affected the thermal conductivity greatly. Fig. 1 was the results of self-sintered carbon plate graphitized at 2600ºC without catalytic. It showed that with the increasing of oxidization temperature, the thermal conductivity increased to a maximum value, then decreased. In this case, the highest thermal conductivity was 852w/mK at oxidization temperature of 260°C. Fig. 2a was SEM of carbon plate supported boron catalyst and Fig. 2b was SEM of carbon plate after boron catalytic graphitization. From Fig. 2, we can found that boron mainly in the form of nano-size H_3BO_3 attached to the surface of carbon plate, but it disappeared after graphitization. Boron catalytic graphitization results were showed in fig. 3 and table 1. With the increasing boron content both thermal conductivity and graphitization degree increased to a maximum value, then decreased. The optimal boron content was 3wt%, and at this point the conductivity was 992.4w/mK, graphitization degree was 99.77%. Titanium catalytic graphitization results were showed in fig. 4 and table 2. With the increasing titanium content both thermal conductivity and graphitization degree increased to a maximum value, then decreased. The optimal titanium content was 3wt%, and at this point the conductivity was 922.0w/mK, graphitization degree was 80.46%. The composite catalytic graphitic was showed in table 3 and fig.5. Fig. 5 showed that Ti-B sol soaked carbon plate had a thick film with TiO_2 and H_2BO_3 nano-particle on its surface. After graphitization, some white spot appeared on fiber surface. The highest conductivity of this catalytic system was 996.45w/mK, and graphitization degree was 83.6%.

Table 1 Thermal Conductivity and Graphitization Degree of Boron Catalytic graphitized Carbon Plate

B %	ρ mΩ·cm	λ W/M·K	d_{002} nm	**g %**
0%	0.0716	758.53	3.3757	74.77
2%	0.0333	954.59	3.3707	80.58
3%	0.0276	992.40	3.3542	99.77
4%	0.0285	986.18	3.3558	97.91
5%	0.0343	948.06	3.3608	92.09

Table 2 Thermal conductivity and graphitization degree of the carbon plate after immerging in Ti-sol.

Ti %	ρ mΩ·cm	λ W/M·K	d_{002} nm	g %
0	0.0716	758.53	3.3757	74.77
2	0.0442	889.05	3.3758	74.65
3	0.0385	922.00	3.3708	80.46
4	0.0407	908.08	3.3788	71.16
5	0.0580	818.90	3.3722	78.84

Table 3 Thermal conductivity and graphitization degree of the carbon plate after immerging in Ti-B sol.

No.	ρ mΩ·cm	λ W/M·K	d_{002} nm	g %
1	0.0317	964.80	3.3657	86.40
2	0.0271	996.45	3.3681	83.60
3	0.0277	992.16	3.3683	83.37
4	0.0285	986.25	3.3649	87.32

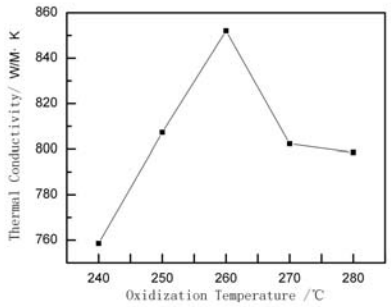

Fig. 1 Thermal conductivity of the carbon plate of different oxidization temperature

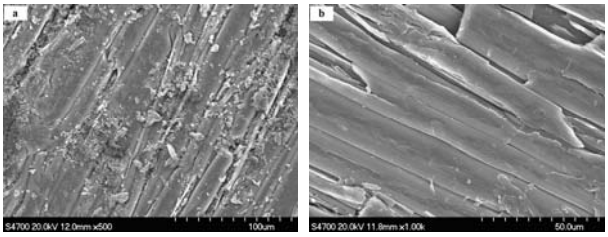

Fig 2 SEM of carbon plate before and after boron catalytic (a: before graphitization; b: after graphitization)

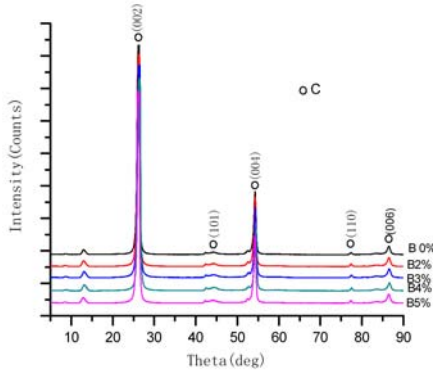

Fig 3 XRD of Boron Catalytic graphitized Carbon Plate

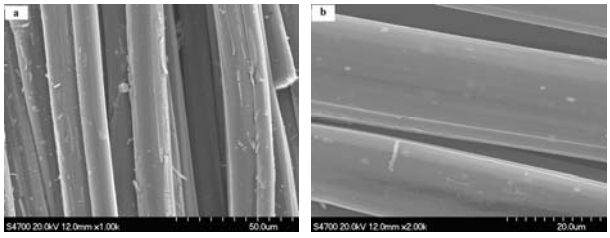

Fig. 4 SEM of carbon plate before and after titanium catalytic graphitization (a: before graphitization; b: after graphitization)

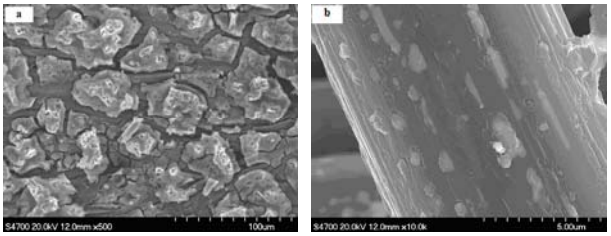

Fig. 5 SEM of carbon plate before and after Ti –B catalytic graphitization (a: before; b: after)

Conclusions

High thermal conductivity of carbon plate can be prepared from mesophase pitch. The oxidation temperature effect the thermal conductivity greatly, and the pitch fiber oxidized at 260°C will obtained highest thermal conductivity carbon plate of 852w/mK. Boron, titanium, and their compound were three catalytic systems for catalytic graphitization. The highest thermal conductivity carbon plate of 996.45w/mK was obtained by Boron, titanium compound catalytic system.

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