EFFECT OF BORON LOADING ON ORIENTATION AND BONDING STRENGTH OF GRAPHITE BLOCK FROM POLYIMIDE FILMS

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Introduction

Highly oriented graphite films are prepared from polyimide (PI) films by heat treatment above 3000°C [1-2]. Boron (B) or rare earth loading on carbon materials accelerates their graphitization [3]. Therefore, authors intend to prepare of highly oriented graphite block by piling up PI films with B and rare earth, and hot pressing. B is expected to accelerate graphitization of PI films and improve bonding strength of films at low temperature. Rare earth loading is expected to improve graphitization and atomic scattering ability of graphite block.

In this paper, graphite blocks were prepared from PI films with or without B by hot pressing. Preparing condition of the blocks was examined in relation to cross sectional texture and crystalline parameters of the graphite blocks. Preferred orientation and bonding strength of the blocks were evaluated using X-ray pole figure method and peeling test, respectively.

Experimental

PI film used was commercial Kapton film with 25 μm in thickness and 15 × 15 mm in size. B was deposited on one surface of the film by PVD method. Amount of B was 0.05 ~ 0.1 mass% as against the film. Twenty five films with or without B were piled up, and set into hot-pressing furnace. The films were carbonized by heating up to 1000°C at a rate of 2°C/min and retaining for 1 hour, and then heating up to 1500°C at the same rate and retaining for 1 hour in *vacuo*. After carbonization, the films were graphitized by heating up to 2200°C at a rate of 20°C/min and retaining for 5 hours in Ar gas. During heat treatment, three patterns of pressing were applied: 1) all pressing; pressing of 67 MPa during treatment, 2) non pressing; no pressing during treatment except for weight of jig and 3) partially pressing: pressing of 67 MPa for 1 hour after retraining 4 hours at 2200°C.

X-ray diffraction patterns were obtained for pulverized blocks with standard silicone by the using of CuKα radiation. Interlayer spacing, d_{002} was measured from the 002 diffraction profile. SEM observation was performed for polished cross section of the graphite blocks. Peeling test was performed for the graphite block sandwiched by two aluminum plates with glue by tensile tester. Peeling strength of the graphite block was calculated by dividing the area of the peeling plane into peel load. The pole figure of 002 plane parallel to the surface of the graphite block was measured.

Results and Discussion

In general, the PI film shrank by heat-treatment, and then it elongated in the direction parallel to the film surface over 1500°C, due to graphitization [1]. The shrinkage of the all-pressed sample without B was 19% ~ 24%, and that of the non-pressed sample without B was 13% ~ 16%. Because of the treatment is the same condition up to 1000°C, the shrinkage of all the samples must be same at 1000°C. Therefore, the all-pressed samples were restrained in elongation. This suggested that pressing between 1500°C to 2200°C restrains graphitization.

The interlayer spacing, d_{002} is shown in **Fig. 1**. The B loaded sample showed lower value of d_{002} than that of the B free sample. The all-pressed sample showed higher value of d_{002} than that of the other samples.

Several narrow gaps were observed in the all-pressed and partially-pressed samples, while some large gaps were observed in the non-pressed sample by SEM observation. Some large gaps like elliptical pore were observed in the all-pressed samples with B. The amount of gaps, however, decreased in the non-pressed and partially-pressed samples with B comparing with the B free samples. Especially, the partially-pressed sample had no gaps and pores as shown in **Fig. 2**. This suggested that pressing and B loading decrease the gaps of the graphite block from PI films. The peeling strength of the block from PI films with or without B is shown in **Table 1**. Peeling strength of the pressed samples was

higher than that of the non-pressed sample, and that of the B loaded sample was much higher than that of the B free sample.

Pole figures of carbon 002 plane of the non-pressed sample with or without B are shown in **Fig. 3**. These are projected figures from broadside of the three-dimensional pole figures. The pole of 002 plane of the non-pressed sample was concentrated on the center of the figure. Pole figures of the all-pressed sample and partially-pressed sample showed that the pole of 002 plane was concentrated on the center of the figures like the non-pressed sample. This suggested that the 002 plane of the block from PI films with or without B oriented almost parallel to the surface of the original films. Half-width of the B loaded sample was smaller than that of the B free sample, and the non-pressed samples showed smaller value of the half-width than the samples pressed by other pattern. Thus, it was suggested that the orientation of carbon 002 plane of graphite block from PI films is improved by B loading, but is debased by pressing.

Conclusions

Graphite blocks were prepared from 25 films of polyimide by hot pressing at 2200°C with or without B. B loading accelerates graphitization and decreases gaps between films. Pressing between 1500°C and 2200°C restrains graphitization. The blocks with B were strongly oriented in 002 plane. Boron loading improves the bonding strength of the block. Pressing debases orientation in 002 plane.

References

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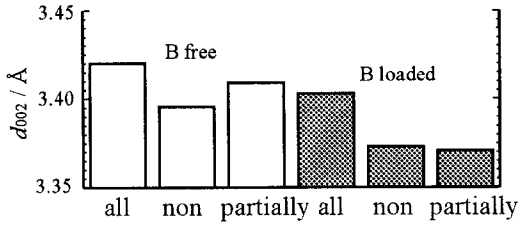


Fig. 1 d_{002} of B loaded and B free graphite blocks heat-treated at 2200°C under three patterns of pressing.

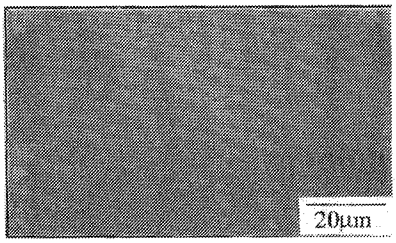
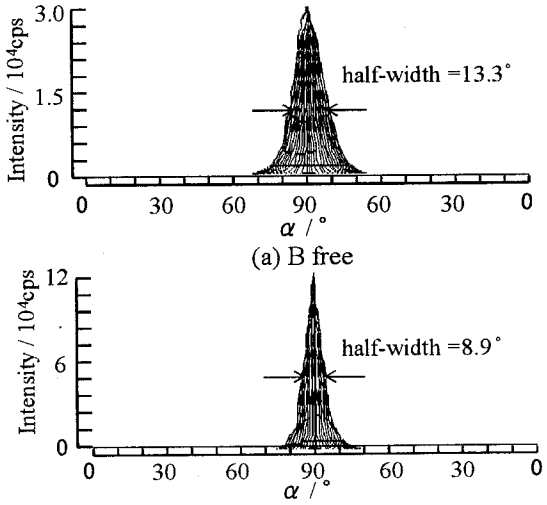


Fig.2. SEM photograph of polished cross section of B loaded graphite block (partially-pressing).

Table 1 Peeling strength (kPa) of graphite blocks from polyimide films

	all	non	partially
B free	63	15	27
B loaded	110	102	166



(b) B loaded

Fig .3 Effect of B loading on 002 pole figure of graphite blocks (partially pressing).

Acknowledgments

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