GRAPHITIZATION OF PETROLEUM COKE WITH CONCURRENT ADDITION OF La_2O_3 AND B IN DIFFERENT La/B RATIOS

Yasuo UCHIYAMA, Shinji ARAKI, Hideaki SANO and Kazuo KOBAYASHI
Department of Materials Science and Engineering, Faculty of Engineering,
Nagasaki University,1-14 Bunkyo-machi, Nagasaki, 852 JAPAN

INTRODUCTION

In previous paper, we reported abrupt change in lattice parameter of petroleum coke with 5 mass% lanthanum hexaboride(LaB_6) with heat treatment temperature[1]. Namely, above 2200°C, LaB_6 caused an expansion in interlayer spacing($d_{(002)}$) and a reduction in lattice size($Lc_{(002)}$) of the coke samples. The phenomenon above 2200°C of the petroleum coke with LaB_6 was similar to that of the natural graphite with LaB_6. A concurrent addition of boron and lanthanum compound has the same effect on the graphitization of the coke powder as a LaB_6 addition. This leads the conclusion that the effect of LaB_6 addition on the graphitization of the coke powder was co-operative effect of boron and lanthanum [2].

In this context, an effect of La/B ratio on lattice parameter of petroleum coke concurrently added with lanthanum oxide and boron was investigated in this paper.

EXPERIMENTAL

Petroleum coke powder was used as a carbonaceous material and lanthanum oxide (La_2O_3) and boron(B) was used as an additive. La_2O_3 up to 1 atom% and B up to 6 atom% was concurrently mixed to the petroleum coke by grinding in alumina motor for 0.5 h. Composition and La/B ratio of the samples was listed in Table 1. The mixed powder was then heat-treated for 1 h at temperatures from 1500°C to 2500°C under an inert gas atmosphere. The heating rate was 20°C/min.

RESULTS AND DISCUSSION

Figure 1 shows $d_{(002)}$ of the petroleum coke with La_2O_3 and B, along with that of the petroleum coke. At 1500°C, $d_{(002)}$ was independent on La- and B-content. At 1800°C, $d_{(002)}$ of the petroleum coke with La/B=1/12 was as large as that of the petroleum coke, while that of the petroleum coke with La/B=1/3 was smaller. In the case of the sample with La/B=1/6, it decreased with an increase in La-content. This means that La accelerated a structural change of the petroleum coke. In the case of the sample with a constant La content of 0.5 atom%, $d_{(002)}$ increased with an increase in B content. This means that B decelerated a structural change of the petroleum coke under La presence, which is contrary to a mono addition of B[3].

Table 1 Composition of sample powder

sample name	composition (atom %)				La : B
	C	La	B	O	
C - 3.31mass% La_2O_3 - 2.64mass% B	96.375	0.25	3.00	0.375	1 : 12
C - 6.44mass% La_2O_3 - 1.28mass% B	97.25	0.50	1.50	0.75	1 : 3
C - 6.44mass% La_2O_3 - 2.57mass% B	95.75	0.50	3.00	0.75	1 : 6
C - 6.46mass% La_2O_3 - 5.15mass% B	92.75	0.50	6.00	0.75	1 : 12
C - 12.2mass% La_2O_3 - 2.44mass% B	94.50	1.00	3.00	1.50	1 : 3
C - 12.3mass% La_2O_3 - 4.89mass% B	91.50	1.00	6.00	1.50	1 : 6
C - 3.99mass% La_2O_3 - 1.59mass% B	97.42	0.3035	1.822	0.4553	1 : 6

At 2000°C, $d_{(002)}$ of the sample with La_2O_3 and B was smaller than that of the petroleum coke, as shown in Fig. 2. $d_{(002)}$ of the sample with large La/B ratio was large than that of the sample with smaller La/B ratio. At this temperature, B was more effective on the structural change of the petroleum coke.

Above 2200°C, on the other hand, $d_{(002)}$ of the sample with La/B=1/3 or 1/6 was larger than that of the petroleum coke, while that of the sample with La/B=1/12 was smaller, as shown in Fig. 3. At 2200°C and above, B accelerated the structural change of the petroleum coke as usual[3], and abrupt change in lattice parameter occurred in the sample with large La/B ratio.

$Lc_{(002)}$ was below 100 angstrom and independent of La- or B-content at 1500°C. Above 1800°C, although it complicatedly depended on La- or B-content, it had a tendency to be larger as $d_{(002)}$ decreased.

CONCLUSIONS

Structural change of petroleum coke concurrently added with lanthanum oxide and boron depended on La/B ratio as well as on heat treatment temperature. Abrupt change in lattice parameter occurred in the sample with large La/B ratio above 2200°C.

REFERENCES

[1] Y. Uchiyama, S. Araki, H. Sano and K. Kobayashi, Carbon, **32**, 1388(1994).

[2] Y. Uchiyama, S. Araki, H. Sano and K. Kobayashi, Carbon, **33**, in press(1995).

[3] K. Kobayashi, H. Funahashi, H. Sano, H. M. Cheng and Y. Uchiyama, TANSO, **1993**, 15(1993).

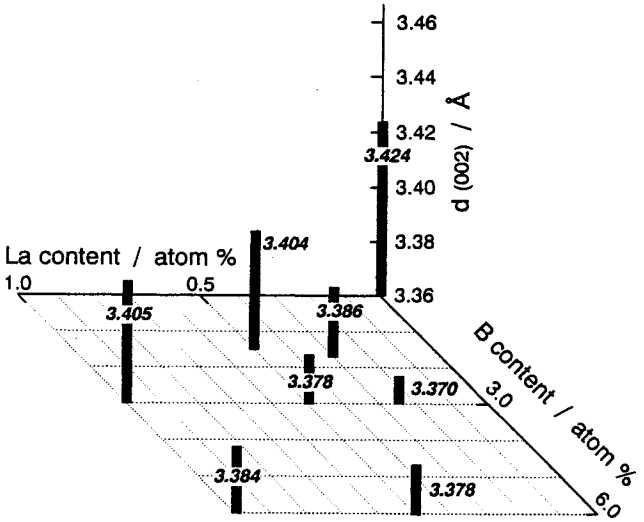

Fig. 2 Effect of La- and B-content on d (002) of the 2000°C -treated petroleum coke with La_2O_3 and B.

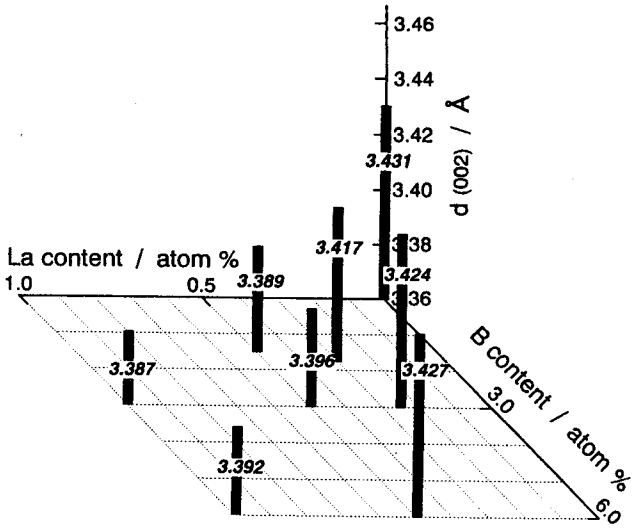

Fig. 1 Effect of La- and B-content on d (002) of the 1800°C -treated petroleum coke with La_2O_3 and B.

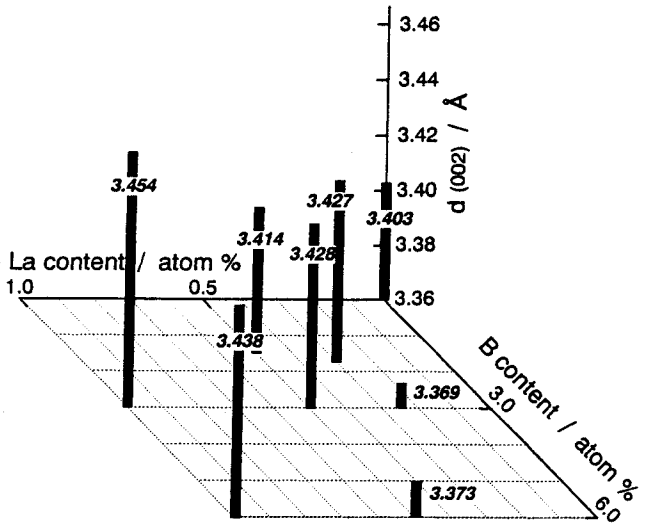

Fig. 3 Effect of La- and B-content on d (002) of the 2200°C -treated petroleum coke with La_2O_3 and B.