EFFECT OF HOT-PRESSING TEMPERATURE ON SINTERED POLYCRYSTALLINE GRAPHITE MADE FROM PITCH COKE WITH B_2O_3 ADDITION

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Introduction

K. Kobayashi et al[1] have reported that high dense and strong polycrystalline graphite was fabricated by hot-pressing method from calcined pitch coke powder with B_2O_3 addition. In that case about 10% B_2O_3 addition was found to be most effective for sintering and graphitization of coke powder.

This paper describs on effect of hot-pressing temperature influenced on some properties of polycrystalline coke compacts made from calcined pitch coke powder with 10% B_2O_3 addition at various temperatures from 1200 to 2200°C under a pressure of 200kg/cm^2 comparing the compacts made from same powder with no addition under same condition.

Experimental

Both calcined pitch coke powder with 10wt% B_2O_3 added as aqueous solution and the powder with no addition were used as the starting materials. After drying the samples in a drying oven each 15g was set in a graphite die of 30mm in diameter and hot-pressed at various temperature from 1200 to 2200°C under a pressure of 200kg/cm^2. The holding time at the desired temperature was set for 30 min and after cutting off the heating the sample was kept within a die for 15 min under the pressure.

After hot-pressing the compacts of 30mm in diameter and 10-14mm in height were obtained. In order to know the effect of hot-pressing temperature, density, mechanical strength, graphitization degree, boron and boron carbide content of the compacts were examined.

Results and discussion

Fig.1 gives change of bulk density of the compacts with hot-pressing temperature. The compacts from the powder with B_2O_3 addition shows almost constant value of 1.6g/cm^3 in the range from 1200 to 1800°C and then the value increase to about 2.0 g/cm^3 at 2200°C. While the compacts from the powder with no addition shows continuous increase with increase of hot-pressing temperature, but the values are all lower than those from the powder with B_2O_3 addition.

Fig.2 gives change of bending strength and compressive strength of the compacts with increase of hot-pressing temperature. The strength once decrease with increase of the temperature, shows minimum value at 1600-1800°C and raise again above 2000°C in the case of the compacts from the powder with B_2O_3 addition. While strength of the compacts from the powder with no addition increase slightly with rise of the temperature, but the value is very small even at 2200°C. It is interesting to notice that both kinds of compacts have almost same strength at 1600-1800°C range.

Fig.3 gives change of d(002) spacing of coke with hot-pressing temperature. From 1200 to 1800°C both compacts from B_2O_3 addition and no addition show same values at the temperature range from 1200 to 1800°C, but at 2000 and 2200°C d(002) spacing of the compacts from the powder with B_2O_3 addition is considerably lower than those from no addition. The value of 3.356A at 2200°C is nearly equal to that of natural graphite. Apparent crystallite size, Lc(002), increased remarkably above 2000°C comparing that of coke powder with no addition as shown in Fig.4. (112) reflection peak of the compact from the powder with B_2O_3 addition was also observed to develop above 2000°C. From these X-ray data it is known that graphitization is accelerated by B_2O_3 addition above 2000°C.

Fig.5 gives change of total boron content and content of boron forming as boron carbide structure with increase of hot-pressing temperature in the compacts made from the powder with B_2O_3 addition. Total boron content is almost constant about at 2.5wt%. Boron carbide is formed at 1600°C and the content decrease with increase of the temperature. In the range from 1200 to 1400°C boron exists as a form of boron oxide and in the range from 1600 to 1800°C most of boron exite as a form of boron carbide structure. Above 2000°C a part of boron is considered to solid-soluted into coke structure and promote sintering and graphitization of coke.

It is concluded that the hot-pressed compacts made from coke powder with 10wt% B_2O_3 addition could be classified into three types. The first is the one made below 1400°C in which coke particles adhered each other with B_2O_3 glass phase, the second is the one made between 1600 and 1800°C which consist of the mixture of independent coke particles and small amount of boron carbide, and the last is the one made above 2000°C of which boron is considered to give strong influence for sintering and graphitization of coke by its diffusion into coke structure.

Therefore in this experiment hot-pressing temperature above 2000°C is found to be necessary for obtaining dense and strong polycrystalline graphite.

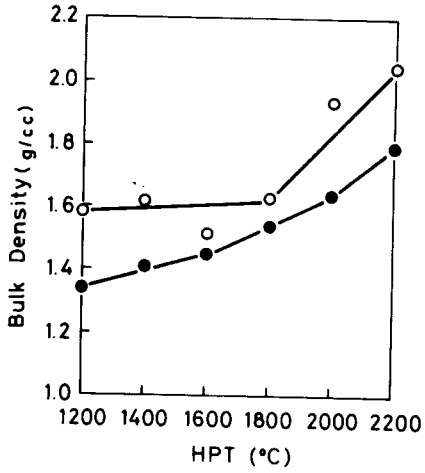
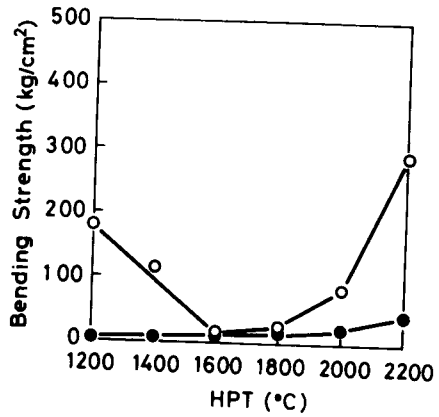


Fig.1 Change of bulk density of the compacts with hot-pressing temperature.
○: coke with 10% B_2O_3 addition
●: coke with no addition



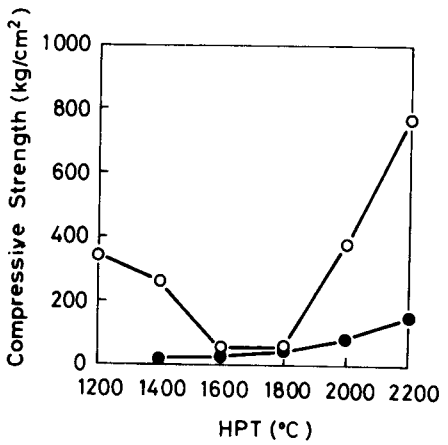


Fig.2 Change of bending strength and compressive strength of the compacts with hot-pressing temperature.
○: coke with 10% B_2O_3 addition
●: coke with no addition

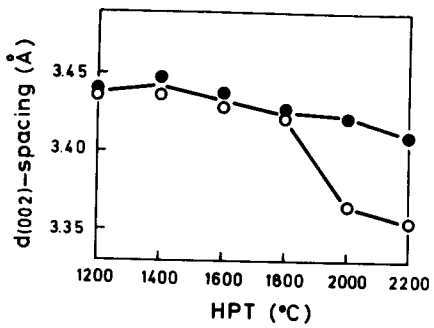


Fig.3 Change of d(002) spacing of coke with hot-pressing temperature.
○: coke with 10% B_2O_3 addition
●: coke with no addition

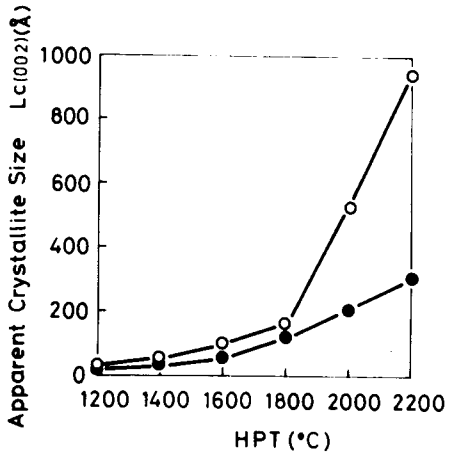


Fig.4 Change of apparent crystallite size, Lc(002), of coke with hot-pressing temperature.
○: coke with 10% B_2O_3 addition
●: coke with no addition

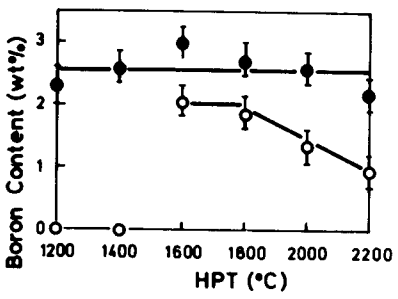


Fig.5 Change of boron content in the compacts with hot-pressing temperature.
●: total boron content by chemical analysis
○: content of boron forming B_4C estimated by X-ray analysis

Reference

(1) K. Kobayashi et al, Abstract for the 12th Carbon Conf. (1975)