Article ID: 6326(1999)03 - 0477 - 04

Synthesis of high grade octahedron diamond through epitaxial growth[①]

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Abstract: The work was based on the relationship between the crystal structure and parameters of diamonds in the synthetic process, in which Ni-Mn-C powder catalyst was used. At first, the diamond crystals nucleated in a cubic-octahedron morphology through controlling such synthetic parameters as temperature and pressure in a parameter region where cubic-octahedron ctystals can be obtained. Then the diamond nuclei grew epitaxially into crystals with a perfect octahedron morphology through increasing the synthesis temperature and decreasing the synthesis pressure in a parameter region where octahedron crystals can be obtained.

Key words: high grade; diamond; eqitaxial growth **Document code: A**

1 INTRODUCTION

The diamond synthesized at isostatically high pressure and high temperature with the action of catalyst belongs to the cubic crystal system, while there are several kinds of crystal structures of common synthetic diamond, such as cubic, octahedron, cubic-octahedron and irregular cryatal structures, The crystal structure of diamond has a great influence on its application and evaluation, and has drawn much attention of superhard material scientists[1~6]. Previous research showed that, there is a relationship between the crystal structure of diamond and the synthesis temperature and pressure; in the whole parameter region where diamond can be synthesized, there are several different small regions corresponding to the synthesis of diamonds with different crystal structures, shown as Ref.[7].

Most of the domestic synthetic diamomds using NiMnCo catalyst have a cubic-octahedron crystal structure[8~10]. Luo *et al*[3] had studied the synthesis of high grade diamonds, and had produced diamonds with an octahedron crystal structure and sharp blades. Because of the high potential of octahedron diamonds in application, their work had draw much attention[11]. However, in current references[1,12~13], the octahedron diamonds reported had incomplete crystal structure, and no such diamonds were synthesized through direct synthetic process.

This work adopted Ni-Mn-C catalyst, and aimed at experimentally producing high grade octahedron diamond through the epitaxial growth of the cubic-octahedron diamond nuclei into octahedron crystals.

2 EXPERIMENTAL

2.1 Raw materials

Ni-Mn-C powder catalyst with a fish-scale appearance[14] was used. The T64P type graphite powders were produced in Dongxin Electric Carbon Works (Zigong, Sichuan povince), their graphitization degree was 90%, The cleavelandite was produced in Mentougou, Beijing.

2.2 Synthesis at high pressures and high temperatures

The Ni-Mn-C catalyst powders and the T64P graphite powders were mixed, and then compacted to cakes at a pressure of 1 MPa. The cleavelandite powders were also pressed to neces-

① Received Oct.8, 1998; accepted Eeb.12, 1999

sary shapes, and then the cleavelandite compacts were installed after being calcined at 400 ℃

The synthesis of diamond was conducted in a DS6 × 800A type diamond-synthesizing press. The compacts were put in the cleavelandite chamber layer by layer, as shown in Fig.2.

Fig.1 Schematic diagram of synthesis of diamond

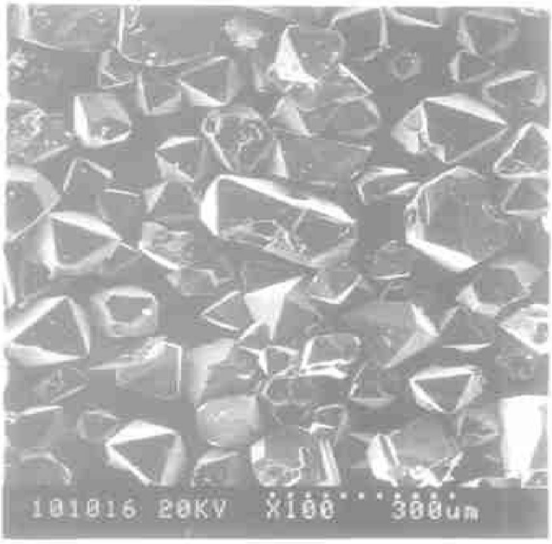

Fig.2 Morphology of diamonds synthesized through slightly increasing temperature

The synthetic parameters were refered to that in reference [3]. In order to avoid the occurrence of diamonds with other crystal structures, the synthetic parameters were firstly controlled in the parameter region where cubic-octahedron diamond can be produced. Therefore, then they were changed to the parameter region where octahedron diamond can be produced. Therefore, cubic-octahedron diamond nuclei were firstly formed, and then they grew epitaxially into octahedron crystals.

3 RESULTS

3.1 Octahedron diamond synthesized by increasing temperature ($\Delta T > 0$)

After being nucleated in the cubic-octahedron crystal region, the octahedron diamonds prepared by simply increasing temperature are shown in Fig.3 and Fig.4. No crystal structural change occurred when $\Delta T < 10$ ℃, as shown in Fig.3. However, dissolution of octahedron crystal occurred at $\Delta T = 50$ ℃, as shown in Fig.4. The probable reason lies in that the synthetic parameters had surpassed the graphite-diamond equilibrium region, and the reverse reaction of diamond → graphite occurred.

Fig.3 Morphology of diamonds synthesized through largely increasing temperature

3.2 Octahedron diamond synthesized by decreasing pressure ($\Delta p < 0$)

Decreasing pressure is another method for the transition of synthetic parameters from cubic-octahedron region to the octahedron region. Fig.5 shows the synthesized octahedron diamond at $\Delta p = -0.2$ MPa. In our experiments, it is difficult to obtain octahedron diamonds by simply decreasing pressure. Therefore, the synthetic parameters rigion for the growth of octahedron

Fig.4 Morphology of diamonds synthesized through decreasing pressure

Fig.5 Morphology of octahedron diamond of high grade

diamond is narrow.

3.3 Synthesis of high grade octahedron diamond

In order to decrease the change of pressure and temperature in the synthetic chamber, the increase of temperature and the decrease of pressue were conducted alternatively, there fore, the synthetic parameters slowly transitted to the octahedron crystal region. After holding for 300 s, it is found that the diamond had grown epitaxially into an octahedron crytal with a perfect shape and complete {111} crystal planes, as shown in Fig.5.

4 DISSCUSION

4.1 A model for epitaxial growth of octahedron diamond

According to the P-T diagram for diamond synthesis[7], the growth rates of {111} and {100} cyrstal planes in different synthetic regions are different. In high temperature synthetic region, {111} were the preferential growth planes, while in high pressure synthetic region, {110} were such planes that when the cubic-octahedron nuclei were treated in the octahedron crystal region, the growth of {100} crystal planes were restrained, while {111} crystal planes grow preferentially. Therefore, perfect octahedron diamonds were obtained through the complete growth of {111} crystal planes, as shown in Fig.6.

4.2 Control of crystal structure in the synthesis of octahedron diamonds

As shown in Fig.1, the octahedron crystal region is in a high temperature area which is closely adjacent to the graphite-diamond equilibrium line, so the octahedron crystal structure is also called "high temperature crystal structure". Because the cleavelandite will tranformed to coexite and cyanite in the synthetic process at the expense of the chamber pressure[15], the synthesis of octahedron diamond should be conducted at a high pressure to compensate the loss of the pressure. In the meantime, the coarsening of diamond single crystal should be avoided because the connection of "metal film" on the diamond surface will lead to the decrease of the resistance of the synthetic bar and the increase of the temperature.

In this work, the size of the synthesized diamonds was below 300μm, and the synthesis of large-sized diamonds need further study.

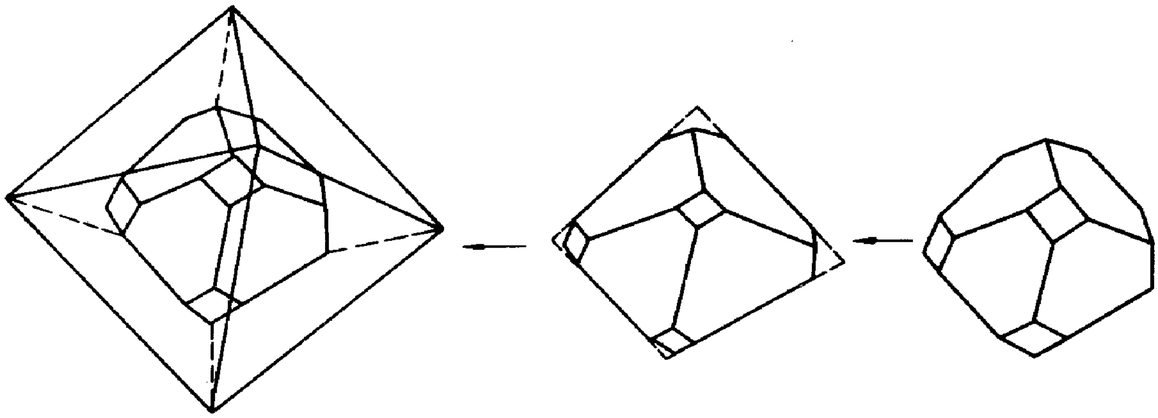

Fig.6 Epitaxial growth of octahedron diamonds

5 CONCLUSIONS

The control of diamond crystal structures in this work was proved to be effective. Through changing the synthetic temperature and the synthetic pressure alternatively, the cubic-octahedron diamond nuclei grow epitaxially into octahedron diamond crystals, and high grade diamonds can be obtained.

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(Edited by Zhu Zhongguo)