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Synthesis and characterization of polycrystalline sintered compacts of cubic boron nitride

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Synthetic cubic boron nitride (CBN) compacts are an important tool material used extensively for the machining of hardened steels. The paper describes work on the synthesis and characterization of CBN compacts. A 200 tonne cubic press has been used for the generation of high pressures. Solid solutions of TiN and TiC have been used as the binder material. The CBN powder and the binder are homogenously mixed and the mixed powder is pressed in a steel die under a pressure of about 3 kbars (300 MPa). The pellets so formed are the starting material for synthesizing the compacts. Compacts both with and without a tungsten carbide substrate have been synthesized. The best compacts are formed at 58 kbar (5.8 GPa) and 1450° C. The sintered compacts after grinding and polishing are characterized by using powder X-ray diffraction, scanning electron microscopy and energy-dispersive X-ray analysis. Knoop hardness measurements made on the best CBN compacts give a hardness of 3400 $HK_{0.5}$.

1. Introduction

Polycrystalline sintered compacts of cubic boron nitride (CBN) are an industrial raw material of considerable importance for the machine tool industry. CBN has a hardness next only to diamond. Also, it is thermally stable up to a temperature of about 1000° C and is chemically inert to iron. These properties make CBN compact cutting tools specially suitable for machining nickel-base and cobalt-base superalloys and chilled cast iron.

CBN compacts are synthesized by mixing a binding material with CBN powder, pressing the mixture in the form of a pellet and subjecting the pellet to a high pressure–high temperature environment. The synthesis of a tough, coherent, high strength and stable CBN compact was first reported by Wentorf and co-workers [1]. The binding materials were chosen from the group consisting of tungsten, nickel, beryllium, rhenium, titanium, zirconium, chromium, cobalt, Al_2O_3, BeO, molybdenum, manganese and copper. Hibbs and Wentorf [2] later synthesized CBN compacts on a WC–Co substrate. Fukunaga *et al.* [3] have also reported the synthesis of a sintered compact of CBN–Co–Al on a WC–Co substrate.

This paper describes the work carried out by the authors at the National Physical Laboratory, New Delhi on the synthesis of CBN compacts with or without a WC–Co substrate using solid solutions of TiN/TiC in different weight proportions as the major binding material. The synthesized compacts were characterized using powder X-ray diffraction for phase identification and scanning electron microscopy (SEM) for microstructure and particle size analysis. Energy-dispersive X-ray analysis (EDX) was used for elemental analysis. Knoop hardness measurements on the top and bottom surface of the compacts were made under a 0.5 kg load. The results of these characterizations are also reported in this paper.

2. Experimental details

2.1. Equipment used

High-pressure sintering of the samples was carried out in a 200 tonne hydraulic press of the cubic design shown in Fig. 1. The press is capable of generating a pressure of 70 kbars (7 GPa) in a pyrophyllite cubic capsule of 16 mm edge. A through-hole of 6 mm diameter was drilled in the capsule to contain the experimental materials. The samples were heated indirectly by passing a heavy current drawn from a low-voltage a.c. source through a graphite sleeve surrounding the sample. A high-current step-down transformer provided the low-voltage a.c. source. The current was regulated by an electronic controller which provided a precise control of the heating power.

2.2. Starting materials

CBN powder of 6 to 9 μm particle size (obtained from General Electric, USA) was homogeneously mixed with a solid solution of TiN/TiC of particle size 3 to 9 μm and a small quantity of aluminium powder of particle size less than 1 μm. The mixing was facilitated by the addition of a small quantity of methanol which was later removed by heating the powder to 200° C. The powder mixture was pressed into a disc in a carver press using a steel die and punch and a pressure of about 3 kbars (300 MPa). In a similar manner the substrate material consisting of WC with 9 wt % Co content and particle size 2 to 5 μm was also pressed in the form of a disc. The sizes of the CBN and WC–Co discs for compacts synthesized on a substrate were as follows:

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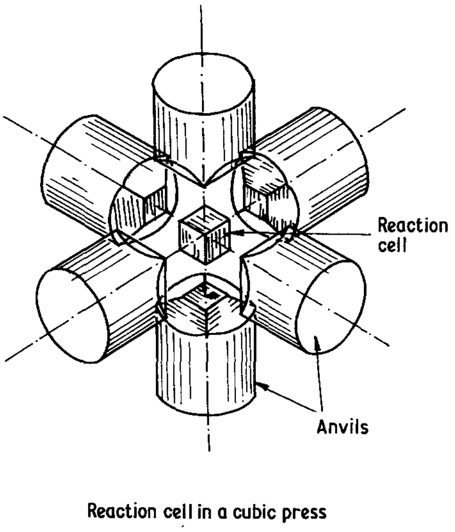

Figure 1 200 tonne cubic press.

TABLE I Details of experiments

Series	Composition (wt %)				
	CBN	TiN/TiC			Al powder
		30:70	50:50	70:30	
A	45	45	–	–	10
	45	–	45	–	10
	45	–	–	45	10
B	50	45	–	–	5
	50	–	45	–	5
	50	–	–	45	5
C	45	50	–	–	5
	45	–	50	–	5
	45	–	–	50	5

CBN : diameter = 6 mm, thickness = 1.5 mm
WC–Co : diameter = 6 mm, thickness = 2.0 mm

For unbacked compacts the dimensions of the CBN discs were as follows:

diameter = 6 mm, thickness = 3.5 mm

The CBN and WC–Co discs as well as the graphite sleeves and pyrophyllite capsules were heated at 300° C overnight to drive out any moisture. The composition of the filled pyrophyllite capsule is shown in Fig. 2. Fig. 2a shows the reaction cell assembly for compacts on a substrate and Fig. 2b the cell assembly for unbacked compacts.

2.3. Experimental procedure

Three series of experiments termed A, B, C and three experiments in each series with different proportions of CBN powder and the binding materials as shown in Table I were performed.

The filled capsule was mounted on the press and the pressure was slowly raised to the desired value. The sample was then heated by passing current through the graphite heater. The desired temperature was attained in 15 min by controlling the electric power fed into the graphite heater. The rate of increase of temperature was about 100° C min^{-1}. The temperature of the sample was measured by inserting Pt–Pt 10% Rh thermocouple wire at the centre of the capsule. The temperature calibration was repeated for each series of experiments. After the sintering was over, the temperature was slowly decreased to room temperature while the pressure was maintained constant. The pressure was then slowly released.

2.4. Optimization of process parameters

Preliminary experiments indicated that satisfactory compacts were synthesized at a pressure of 58 kbars (5.8 GPa). Keeping the pressure constant at this value, a number of experiments were performed to determine the optimum temperature. The temperature was varied in the range 1350 to 1550° C and the sintering time was kept at 20 min. It was found that between 1450 and 1550° C there was partial conversion to hexagonal boron nitride (HBN).

Quantitative X-ray analysis showed that at 1500° C the conversion of CBN to HBN was about 40% by weight. At 1450° C the compacted discs did not contain any HBN. Best compacts were, therefore, synthesized at 58 kbar and 1450° C.

3. Characterization of compacts

3.1. X-ray characterization

Powder X-ray diffraction studies were carried out on

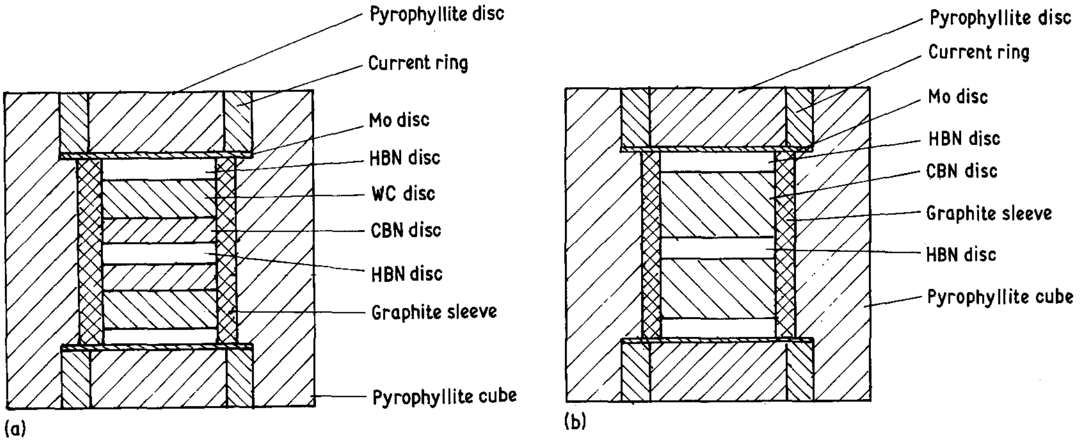

Figure 2 Pyrophyllite capsules (a) for compacts on a substrate, (b) for unbacked compacts. Dimensions (mm) as follows: current ring o.d. 10.0, i.d. 7.4, thickness 3.0; pyrophyllite disc diameter 7.4, thickness 3.0; graphite sleeve o.d. 8.0, i.d. 6.0, length 10.0; molybdenum disc diameter 10.0, thickness 0.25; hexagonal boron nitride (HBN) disc diameter 6.0, thickness (a) 1.0, (b) 1.5; WC disc diameter 6.0, thickness 2.0; CBN disc diameter 6.0, thickness (a) 1.5, (b) 3.0.

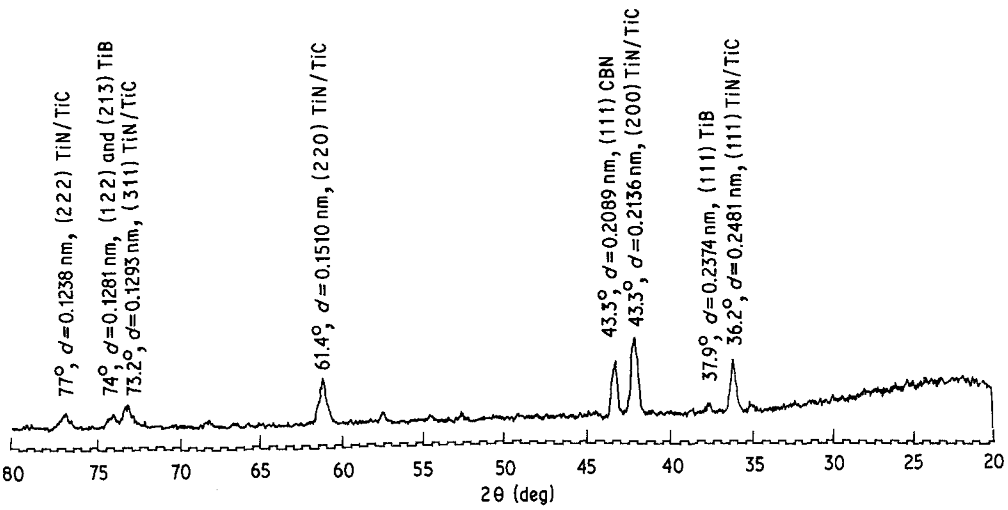

Figure 3 X-ray diffractogram for the CBN compact.

the synthesized compacts for phase identification. $\mathrm{Cu}K_{\alpha}$ radiation was used as the exploring X-ray beam. Fig. 3 shows a typical powder X-ray diffraction pattern. It is seen that the diffraction line due to the HBN which occurs at $2\theta = 26.7°$ is absent. The (1 1 1) diffraction line of CBN is observed at $2\theta = 43.3°$ ($d = 0.2089$ nm). The diffraction maxima observed at $2\theta = 36.2°$ ($d = 0.2481$ nm), 42.3° ($d = 0.2136$ nm), 61.4° ($d = 0.1510$ nm), 73.2° ($d = 0.1293$ nm) and 77° ($d = 0.1238$ nm) correspond to (1 1 1), (2 0 0), (2 2 0), (3 1 1) and (2 2 2) reflections of TiN/TiC solid solution. The observed diffraction maxima at $2\theta =$ 37.9° ($d = 0.2374$ nm), 51.9° ($d = 0.1760$ nm) and 61.2° ($d = 0.1514$ nm) correspond respectively to (1 1 1), (3 0 1) and (0 2 0) reflections of TiB. The diffraction line observed at $2\theta = 74°$ ($d = 0.1280$ nm) is due to superposition of (1 2 2) and (2 1 3) lines of TiB. The strongest line (1 0 2) of TiB at $2\theta = 42.2°$ ($d = 0.2140$ nm) overlaps with the strongest reflection of TiN/TiC. From this analysis it can be concluded that the synthesized CBN compact consists of three crystalline phases, namely CBN, TiN/TiC and TiB. No AlN or Al_2O_3 could be detected.

3.2. Microstructure study with scanning electron microscopy

Electron micrographs were recorded in the secondary electron emission mode with 0° tilt. The following broad features were observed in the micrographs:

(i) In most regions CBN particles are held together by TiN/TiC particles.

(ii) In some regions clustering of CBN and TiN/TiC particles is observed. Clusters of CBN particles are bonded by clusters of TiN/TiC particles.

(iii) CBN–CBN bonding is also occasionally observed.

The micrographs were also used to determine the particle size and particle size distribution of CBN and TiN/TiC particles in the compacts. Since the signal-to-background ratio for CBN particles was poor it was not possible to carry out image analysis through a computer-controlled system attached to the microscope. The computations were, therefore, made manually from the recorded micrographs. In doing so the concept of particle size as adopted by the Federation Européenne des Fabricants de Produits Abrasifs (FEPA) was used. According to this concept "the diameter of the circle which completely encloses the particle defines the particle size". Particle size computations were made at 20 different locations in the SEM micrograph and the following maximum and minimum values of particle sizes were recorded:

CBN: Maximum 9.4 μm
Minimum 3.5 μm
TiN/TiC: Maximum 12.2 μm
Minimum 4.5 μm

Table II shows the particle size distribution at these twenty locations.

It is seen from Table II that the particle size ranges of the CBN and TiN/TiC particles are comparable. The characterization results also confirmed that there was no sensible grain growth during the sintering process.

3.3. Elemental analysis

The compacts were subjected to elemental analysis using energy-dispersive X-ray analysis. For this purpose an ISI Super IIIA SEM (Cambridge, UK) in conjunction with spectrum analyser Tracor Northern TN 2700 was used. The operating voltage was 30 kV. A 0° tilt and 16 mm working distance was used. The distance between the specimen and the detector was 30 mm. The spot size was kept constant at its maximum to increase the emitted X-ray signal. A counting

TABLE II Particle size distribution

Particle size range (μm)	Number of CBN particles	Number of TiN/TiC particles
3.5 to 4.5	6	3
4.6 to 5.5	3	1
5.6 to 6.5	4	2
6.6 to 7.5	3	4
7.6 to 8.5	1	7
8.6 to 9.5	3	2
Above 9.5	–	1

interval of 200 sec was employed to count the emitted X-ray photons. Pure aluminium, silicon and titanium were used to record a standard X-ray spectrum. The weight percentages of different elements in the compact were determined quantitatively by comparing the X-ray counts of elements in the compacts with the corresponding values for pure elements. The following elemental composition was observed in the synthesized compact:

Ti	45 wt %
Si	0.2 wt %
Al	2.0 wt %

It will be observed that the weight percentages of titanium and aluminium are not exactly equal to the weight percentages of these elements in the starting materials. This can be explained in the following way. Since the electron beam used to excite the characteristic X-ray lines of the constituent elements in the specimen sees only a small surface area, a micrometre or so, the irradiated area may not be a true representative of the material composition. This is mainly due to the lack of homogeneity of the starting material. This inhomogeneity also leads to hardness variations on the surface of the compact.

3.4. Hardness measurements

Knoop hardness measurements under a 0.5 kg load were made on the synthesized compacts at different points on the top and bottom surfaces. The hardness varied from point to point on the specimen surface for all the compacts. This variation may be due to local microstructural fluctuations. The maximum hardness of 3400 $HK_{0.5}$ was obtained for compacts with the following weight percentages.

(i) 50% CBN, 45% TiN/TiC (30:70), 5% Al
(ii) 45% CBN, 50% TiN/TiC (30:70), 5% Al

4. Conclusions

The investigations reported in this paper lead to the following conclusions:

(i) The best compacts were obtained with weight percentages 50% CBN, 45% TiN/TiC (30:70), 5% Al and 45% CBN, 50% TiN/TiC (30:70), 5% Al.

(ii) The compacts synthesized with these compositions gave a maximum hardness of 3400 $HK_{0.5}$.

Acknowledgements

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