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Experimental study of plastic deformation during sintering of cubic boron nitride compacts

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Abstract

In this work polycrystalline cubic boron nitride compacts (PcBN), prepared with and without binder, were sintered at 8 GPa and 1850°C using different processing times. X-ray diffraction and Raman spectroscopy were used to investigate the kinetics of plastic deformation during sintering. For compacts sintered in short processing times, both Raman and X-ray diffraction linewidths increased, indicating a high plastic deformation of the grains. For longer processing times (30–120 s) the linewidth decreased, tending to the value of the pristine cBN grains. This dependence on the processing time was interpreted as a result of the competition between the plastic deformation of the grains, dominant for short processing times, and the defect annealing process, which becomes significant for longer processing times. When aluminum binder was used for sintering, it was observed that the processing time required to recover the original linewidth is longer, and the maximum broadening of the lines occurs at a longer processing time compared to the case without binder. These results indicated that the binder phase significantly affects both plastic deformation and defect annealing processes.

Keywords: Cubic boron nitrite (CBN); Mechanical properties; Processing; High pressure high temperature

1. Introduction

Cubic boron nitride (cBN) is a synthetic allotropic form of boron nitride obtained under high pressure and high temperature, usually from hexagonal boron nitride (hBN), similarly to the synthesis of diamond from graphite. Another similarity with diamond is the high hardness of cubic boron nitride, responsible for its wide use in cutting tools, mainly for machining hard ferrous materials where diamond is inefficient since it reacts chemically with these materials at temperatures higher than 700°C.

Cubic boron nitride compacts are usually obtained by sintering of cBN powder with binder, under pressure in the range 6–8 GPa, and temperature in the range 1500–2000°C [1]. During the sintering, the plastic deformation of the powder particles occurs simultaneously with the chemical reaction with the binder in a complicated process which is not totally understood [2,3]. The sintering residual stress can be an important parameter related to the behavior of the compacts used as cutting tools.

Voronin et al. [4] investigated the kinetics of plastic deformation for polycrystalline diamond compacts (PCDs), and the results obtained indicated that, for short processing times, the plastic deformation is the dominant mechanism responsible for the sintering of PCD, as evaluated by the broadening of X-ray diffraction lines of the sintered material. For longer processing times, the defect annealing process started to predominate, as indicated by the decrease of the linewidth of the X-ray peaks.

In this work, the kinetics of the plastic deformation of cBN was studied by measuring the linewidth of the (331) X-ray line and of the cBN Raman peak of samples produced using different processing times. The effect of the binder was investigated comparing the results obtained for compacts sintered with and without binder.

2. Experimental procedure

cBN powder of grain size 28/20 μm was obtained from Bakul Institute for Superhard Materials, Kiev,

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Ukraine. Aluminum powder of average grain size of 1 μm was used as the binder, mixed with the sample in a proportion of 5 wt.%.

PcBN compacts were sintered in a high pressure toroidal apparatus [5], at about 8 GPa and a temperature of 1850°C. The pressure was calibrated using the standard bismuth phase transition procedure. The temperature was generated by indirect heating in a graphite heater furnace and the calibration was performed using a Pt/Pt–13%Rh thermocouple.

Initially, the powder was submitted to high pressure, followed by heating, cooling and, finally, pressure release. The processing times were $t=0$, 15, 30, 60 and 120 s, corresponding to the time interval while the sample was heated under high pressure. The particular case '$t=0$ s' corresponds to the pure effect of high pressure, without heating. After sintering, the compacts were approximately 3 mm thick and had a diameter of about 7 mm.

X-ray diffraction was used to investigate the plastic deformation of the grains through the broadening of the line (331) and, also, to analyze the existence of different crystalline phases formed during sintering. The X-ray diffraction measurements were done in a Siemens D500 diffractometer, using Cu Kα radiation in the angular region between 4 and 150°.

The results from Raman spectroscopy were also used to investigate the stress in the compacts after sintering. The measurements were done in a micro-Raman apparatus using the He–Ne laser line as the excitation light.

3. Experimental results and discussion

Fig. 1 shows the diffraction pattern in the region of the (331) line of cBN, for samples sintered without binder for different heating times. Fig. 2 shows the results for the samples sintered with binder. The full width at half-maximum of the (331) peak was calculated as a function of the heating time, as shown in Fig. 3. As can be seen, samples heated during a short time ($t=$ 15 s) exhibited a marked increase in the line broadening for both cases, probably related to the large plastic deformation of the grains (predominant mechanism). The broadening is smaller for samples with binder at $t=15$ s: the presence of the binder decreases the stress gradients in cBN grains.

For the next time interval, namely $t=30$ s, the linewidth decreases considerably for the binder-free samples, probably as a consequence of the onset of an annealing process. However, for the sample with binder, the linewidth continues to increase. This contrasting behavior between the two kind of samples can be explained by the effect of the chemical reaction between aluminum and cBN in the case of samples with binder, giving hard products such as AlN, which produce extra levels of stress for accommodating the new phases. Furthermore, the chemical reaction with aluminum will likely occur first at the defective parts of the cBN grain surface, especially at the exiting dislocations. This can produce a pinning effect, reducing the mobility of the dislocations and preventing annealing. For longer heating time intervals, namely 60 and 120 s, the linewidth for the sample without binder continues to decrease, approaching in 120 s a value very close to that at $t=$ 0 s. However, for the sample with binder, the recovering of the original linewidth is much slower, probably owing to the restricted mobility of the dislocations in the annealing process.

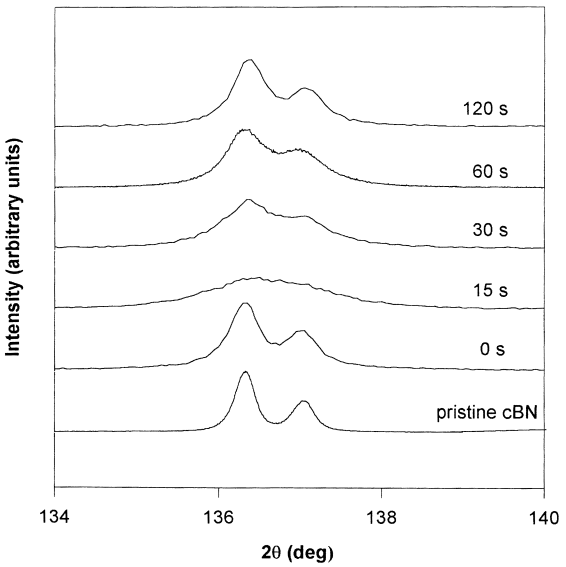

Fig. 1. X-ray diffraction pattern of PcBN sintered without binder for different heating times.

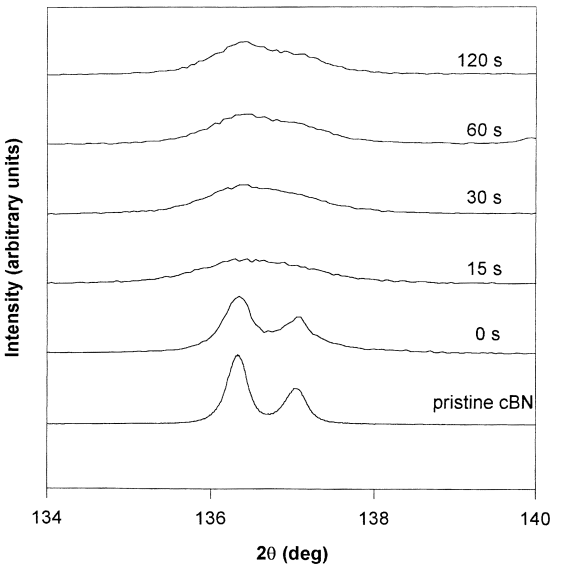

Fig. 2. X-ray diffraction pattern for PcBN sintered with binder for different heating times.

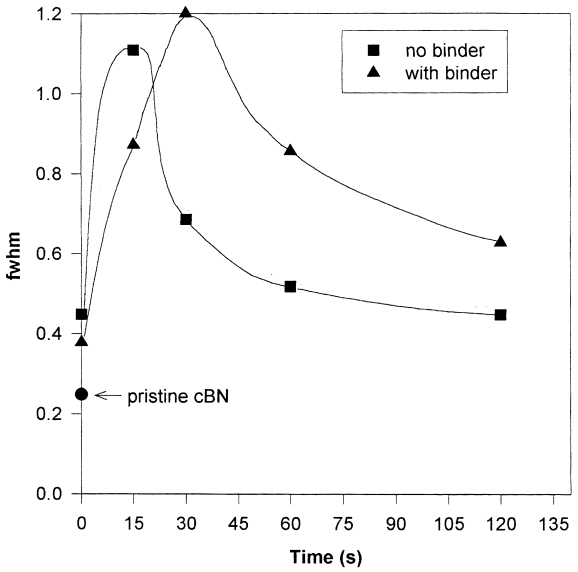

Fig. 3. Linewidth of (331) X-ray line of cBN as a function of the heating time: triangles correspond to compacts sintered with binder; squares correspond to compacts sintered without binder, and the circle represents the pristine cBN linewidth. The solid lines are only for guiding the eyes.

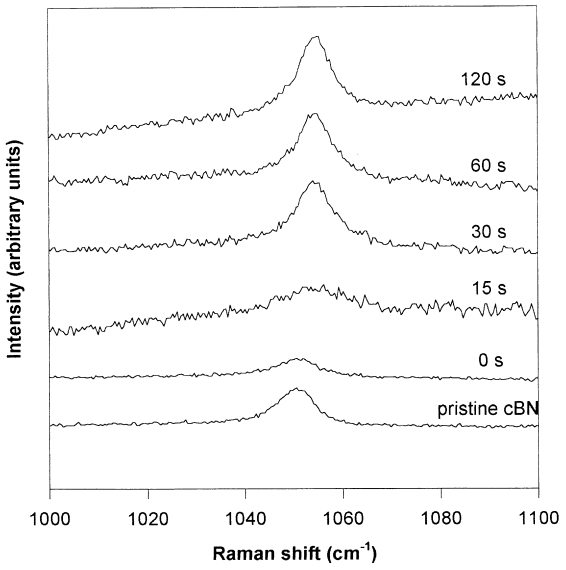

Fig. 4. Raman spectrum of PcBN sintered without binder.

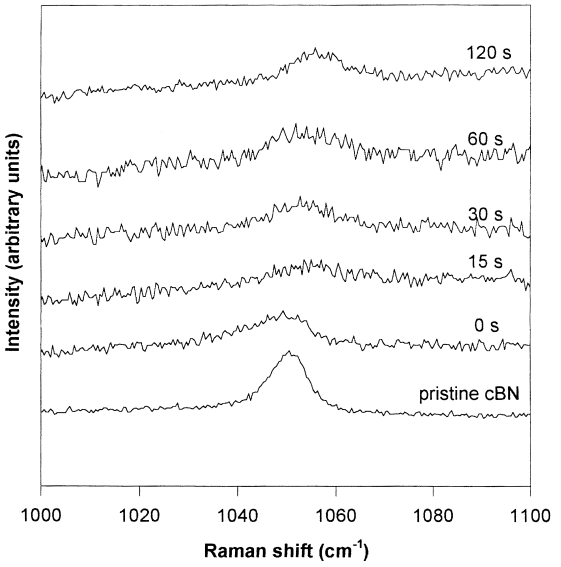

Fig. 5. Raman spectrum of PcBN sintered with binder.

As the heating time increases, the linewidth tends to recover the value obtained for '$t=0$ s', which corresponds to the samples submitted only to high pressure, without heating. As can be seen in Fig. 3, this value for both kind of samples, is higher than the value for the pristine cBN. This pure high pressure stage probably induces the breaking of the original cBN powder grains, reducing their sizes. Comparing the value of the linewidth for '$t=0$ s' in both cases, the samples with binder exhibit a slightly smaller linewidth than the sample with no binder. The binder soft metal phase would reduce the stress concentration at the grain boundaries and, consequently, reduce also the grain breaking process at the high pressure stage.

In the diffraction pattern of the cBN compact sintered with binder in an expanded angular region, there are lines corresponding to cBN and to AlN. It is interesting to note that there is no evidence of the hBN and AlB_2 phases suggested in Ref. [6].

The Raman spectra of the compacts without and with binder are shown in Figs. 4 and 5, respectively. The behavior of the linewidth of the characteristic cBN peak (around 1050 cm^{-1} for pristine cBN) for different heating times is very similar to the behavior of the linewidth of the X-ray spectra. For all samples compacted without heating, there was a broadening of the Raman peak, but there was no peak shift, regardless of the binder. For short heating time, '$t=15$ s', there was an increase in the linewidth of the Raman peak for samples prepared with and without binder and, additionally, a peak shift was observed for both cases. For longer heating times, the linewidth decreased in a way very similar to that represented in Fig. 3 for both kinds of sample. However, the peak shift remained practically unchanged (1053.5 ± 0.5 cm^{-1} for compacts with no binder and 1052.5 ± 0.5 cm^{-1} for compacts with binder). In addition to the intrinsic internal stress level of each sample, this shift may also be related to the grain size: the grain growth is probably smaller for samples sintered with binder owing to the inhibition of the recrystallization of the cBN grains by the presence of the binder. For samples sintered without binder, the grains are probably larger and this may be responsible for the distinct Raman shift.

The density of the compacts, sintered with no binder, was also measured (the interpretation of the density results for samples with binder is complicated owing to

the presence of additional crystalline phases). The density of compacts sintered for $t=15$ s, is 3.42 ± 0.01 g cm^{-3}, which corresponds to 2% porosity. The density of samples sintered for $t=30$–120 s, is 3.47 ± 0.01 g cm^{-3}, which is close to the theoretical value for cBN (3.49 ± 0.01 g cm^{-3}) and corresponds to a porosity of less than 1%. These results show that the densification mainly occurs during the initial stage of sintering as a result of the plastic deformation of the grains. This densification is also responsible for the line broadening of the X-ray and Raman peaks for short time intervals. At longer holding times, when the defect annealing process dominates, there is no substantial increase in the densification.

Hardness measurements in compacts without binder were difficult to make because the grains were not highly bonded. For compacts with binder, sintered with heating, the Vickers hardness results for 200 gf load were in the range 60–70 GPa, but it was difficult to observe a dependence of the hardness on the processing time owing to the low accuracy of the results.

4. Conclusions

In this work we studied the kinetics of plastic deformation in cBN compacts, sintered at high pressure and temperature, with and without aluminum binder. The compacts were analyzed by X-ray diffraction and Raman spectroscopy.

The results obtained indicate that the kinetics of plastic deformation for cBN compacts during sintering is very similar to the kinetics for diamond compacts. For short heating times ($t=15$ s) an intensive plastic deformation of cBN grains was observed, causing a densification of compacts and a broadening of the X-ray and Raman peaks. For short processing times, the plastic deformation is smaller in compacts sintered with binder because it decreases the stress concentration at the cBN grain boundaries. For longer heating times ($t>15$ s) the defect annealing process in cBN compacts dominates, causing a subsequent narrowing of X-ray and Raman peaks. However, the narrowing is much smaller for samples with binder. The interpretation for this behavior is that the presence of the binder generates additional stress by the reaction of Al with cBN. The binder could also reduce the stress relief and the recrystallization rate of cBN grains by hindering the movement of dislocations.

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