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Hot dynamic consolidation of hard ceramics

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Diamond and cubic boron nitride powders were shock compacted at high temperature (873 and 973 K) by using a planar impact system at 1.2 and 2.0 km s⁻¹. Silicon, graphite or a mixture of titanium and carbon powders were added to enhance the bonding of these superhard materials. Hot-consolidated specimens exhibited fewer surface cracks as compared with the specimens shock consolidated at room temperature. Diamond compacts having microhardness values over 55 GPa were obtained by subjecting porous mixtures of diamond crystals (4–8 µm) plus 15 wt % graphite (325 mesh) to an impact velocity of 1.2 km s⁻¹ at 873 K. Well-consolidated c-BN samples, with microhardnesses (starting powders with 10–20 and 40–50 µm) over 53 GPa were obtained.

1. Introduction

It has been shown that shock compaction *per se* is a viable technique for soft materials. However, the high pressures required for compacting hard powders generate tensile stresses which cannot be totally eliminated because of geometrical constraints. These tensile stresses act on the existing flaws and activate them, resulting in cracking. On the other hand, ductile materials require lower pressures; thus the reflected pulse amplitudes are lower. The more ductile materials are also less flaw sensitive. Shock consolidation of extremely hard non-oxide ceramic powders has already been attempted at room temperature by many researchers [1–6]. However, it has been widely reported that there remain two unsolved problems in the shock compaction technique. One is cracking of the compacts at both the microscopic and macroscopic levels. The other is a lack of uniformity in microstructure and mechanical properties within resulting compacts. At three recent workshops held in the United States [7], the Soviet Union [8], and Japan [9], cracking was identified as a major unresolved problem. These two problems tend to multiply with an increase in the shock pressure used. In order to alleviate these two problems, preheating and low shock pressures are desirable. Earlier experiments conducted by Wang *et al.* [10] on nickel-base superalloys indicated that preheating them to 500–700 °C had a very positive effect on the mechanical properties of shock consolidated superalloys. Taniguchi and Kondo [11] and Ferreira *et al.* [12] successfully used higher starting temperatures to help shock consolidation.

Therefore, with the purpose of minimizing the above problems, we tried to consolidate diamond and c-BN powders at high temperatures instead of room temperature. The objectives of this investigation were (1) to preheat the powders to eliminate or minimize cracks in resulting compacts, and (2) to use heat generated from exothermic reaction to help consolidation. In hot shock consolidation, the shock energy

required to melt the powder surfaces is decreased while the powder strength is lower. The effect of particle size on the consolidated diamond and c-BN powders was examined by comparing the results with experiments conducted at room temperature [1–3, 6]. The effect of the addition of graphite, silicon or a mixture of titanium and carbon powders was also examined [2, 4, 13–15].

2. Experimental procedure

Three different sizes of natural diamond powders (4–8, 10–15 and 20–25 µm) and two different sizes of c-BN powders (10–20 and 40–50 µm) were used as starting materials. The composition of the specimens is shown in Table I. The powders were pressed to 60% theoretical density into 5 mm thick and 12 mm diameter stainless steel or Inconel 718 capsules.

The planar impact system developed by Akashi and Sawaoka [16] was used for the consolidation. A schematic illustration of this set-up is shown in Fig. 1a. Twelve capsules can be compacted simultaneously. Fig. 1b shows the set-up for the hot consolidation developed by Yu and Meyers [17]. The planar impact of the flyer plate on the system creates high-amplitude shock waves that transmit through the powders. The detonation is initiated from the detonator at the top of the set-up. The conical lens consists of explosives with two detonation velocities and generates a planar wave in the main charge. The flyer plate is accelerated by the main charge and impacts the capsules, consolidating the powders.

Two experiments were conducted. The capsules containing the powders were preheated to 873 and 973 K and shock compacted at impact velocities of 1.2 and 2.0 km s⁻¹, respectively. The microstructure and crack distribution of the specimens were observed by optical microscopy and scanning electron microscopy, and the crystal structure was analysed by X-ray diffractometry.

TABLE I Experimental conditions for hot consolidation and average Vicker's microhardness values

Composition	Experiment 1: preheat temp. 873 K, imp. vel. 1.2 km s^{-1}		Experiment 2: preheat temp. 973 K, imp. vel. 2.0 km s^{-1}	
	Capsule	Vicker's hardness	Capsule	Vicker's hardness
4–8 μm natural diamond	1-1	29.4	2-1	24.3
10–15 μm natural diamond	1-2	26.0	2-2	27.0
20–25 μm natural diamond	1-3	25.5	2-3	19.6
4–8 μm natural diamond + C (15 wt %)	1-4	> 55.0	2-4	25.8
4–8 μm natural diamond + Si (7.5 wt %)	1-5	15.8	2-5	7.4
10–20 μm c-BN	1-6	37.5	2-6	61.4
40–50 μm c-BN	1-7	53.0	2-7	29.9
40–50 μm c-BN + C (15 wt %)	1-8	24.4	–	–
40–50 μm c-BN + Ti (11 wt %) + C (9 wt %)	–	–	2-9	16.3

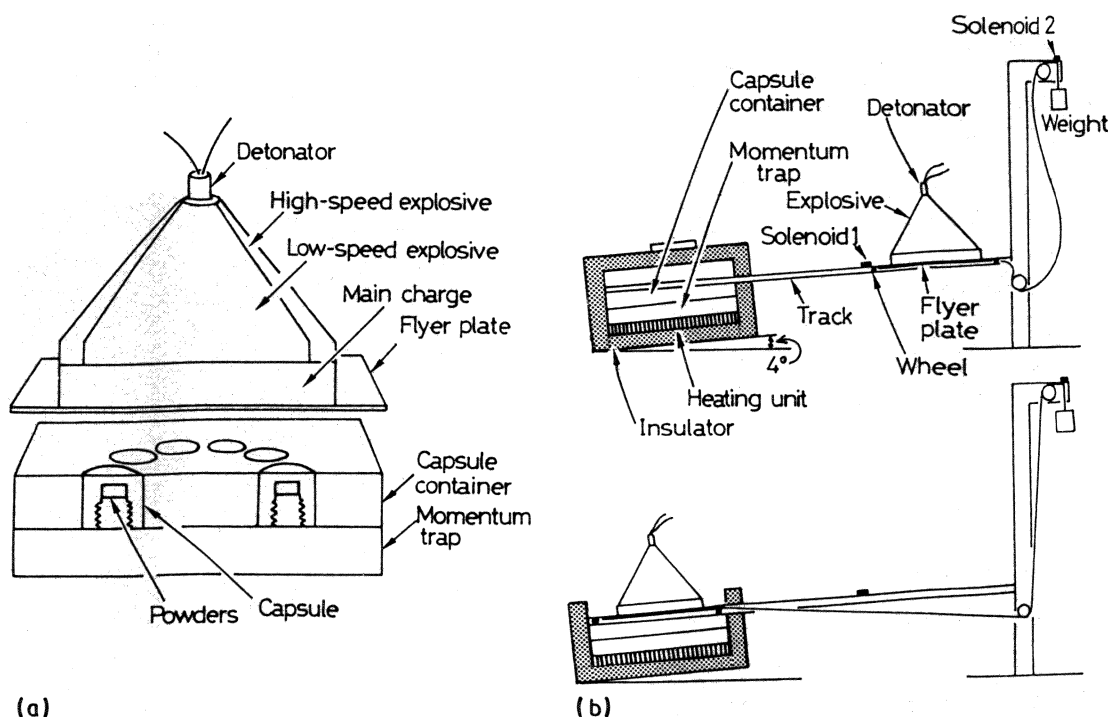


Figure 1 (a) Schematic illustration of planar impact system, and (b) hot consolidation apparatus.

3. Results and discussion

Fig. 2 shows bottom surfaces of diamond + graphite, diamond + silicon and c-BN powders after hot shock consolidation. The bottom surfaces of the specimens shock consolidated at room temperature by Akashi and Sawaoka [5, 6] showed many cracks; cracking is visibly decreased for the high-temperature shock consolidation experiments as shown by Fig. 2a–c. However, cracking is not totally eliminated and circumferential cracks, marked by arrows, are noticeable. High temperature enhances the ease of melting of particle surfaces and decreases the energy required for consolidation. In the case of hot consolidation, flyer plate velocities of 1.2 and 2.0 km s^{-1} were chosen for the consolidation of diamond and c-BN powders, while 1.8 – 3.0 km s^{-1} is required for the consolidation of these powders at room temperature [1–6]. The

decrease of surface cracks is the direct result of the lower shock pressure. The experimental conditions and results are given in Table I. The two shock experiments, at 1.2 km s^{-1} (873 K) and 2 km s^{-1} (973 K), provided two levels of energy delivered to the powders. Experiment 2 delivered considerably higher energy (thermal and shock) than experiment 1; the experiments were planned to establish, to a first order, whether this effect was significant, in the range investigated. In Table I, the first digit represents the experiment number, while the second represents the capsule (e.g. 1-1 is the first capsule in the first experiment). In experiment 1 (1.2 km s^{-1} , 873 K), the capsules containing the specimens were of AISI 304 stainless steel. For experiment 2, the first attempt, using AISI 304 stainless steel capsules, failed because wave focusing produced capsule failure along the top surface of the

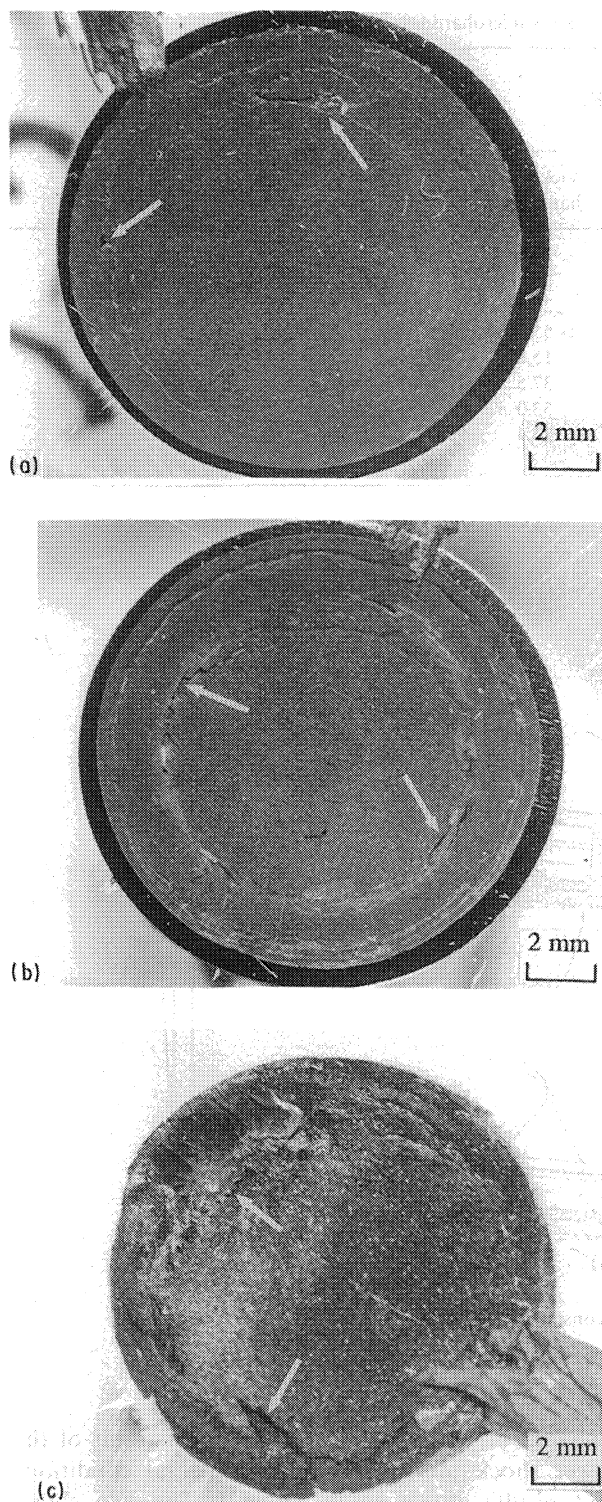


Figure 2 Bottom surfaces of consolidated (a) 4–8 μm diamond and graphite powder mixture, (b) 4–8 μm diamond and silicon powder mixture, (c) 40–50 μm c-BN powders.

disc axis. Therefore, Inconel 718 capsules were used, which had a sufficient strength to resist the combined effects of both high temperature and high stresses.

3.1. Characterization of hot-consolidated diamond powders

Well-consolidated samples were obtained upon shocking diamond powders (4–8 μm particle size) with

60% theoretical density at the impact velocity of 1.2 km s^{-1} . The scanning electron micrograph (Fig. 3) shows that no micro-cracks and voids were observed in this polished surface (capsule 1-1). Small, round, dark areas about 1–2 μm diameter are seen; they could be fractured diamond particles or porosity due to melting and resolidification. This result contrasted with the same sample (2-1) recovered from the second shot in which the hardness appeared to be slightly lower. The higher preheating temperature and impact velocity are not significantly helpful for consolidation of 4–8 μm grade diamond. Fig. 4 shows the cracks that were generated in the consolidated 10–15 μm diamond powder (1-2). The number of cracks generated in the consolidated diamond powders tends to increase with increasing particle size of the starting diamond powders. Generation of small amounts of graphite was detected by X-ray diffraction in the compacted 10–15 μm diamond powders (1-2, 2-2). The transformation to graphite usually occurs in larger particle size specimens consolidated at room temperature [5], but this transformation is enhanced with the



Figure 3 Scanning electron micrograph of polished surface of consolidated 4–8 μm diamond powders (1-1).

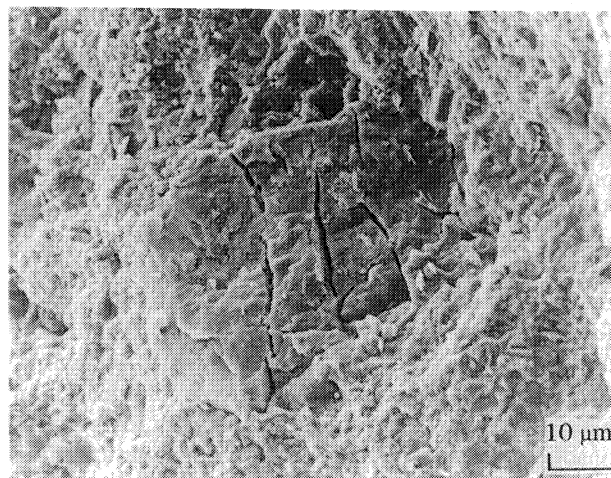
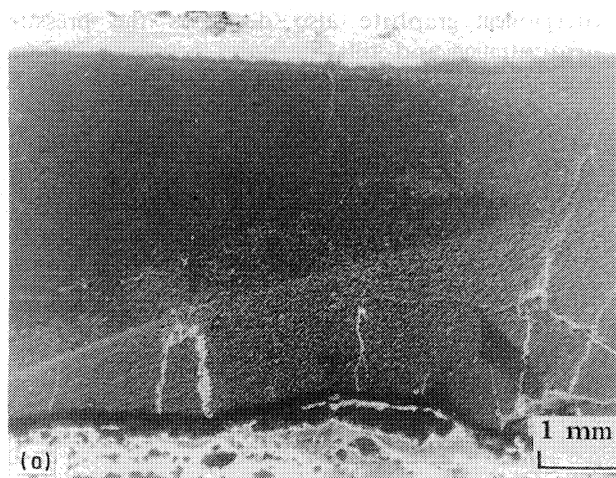


Figure 4 Scanning electron micrograph of polished surface of consolidated 10–15 μm diamond powders (1-2), in which cracks were generated in one particle.

help of preheating. In the case of the hot shock consolidation of pure diamond powders, smaller particle size is required for good bonding, compared with the consolidation at room temperature. Microhardness values in Table I confirm this. Fig. 5a shows half of the vertical cross-section and Fig. 5b shows the Vicker's microhardness distribution of consolidated 20–25 μm diamond powders (1-3). Because the velocity of the shock wave in the powder is considerably lower than in the surrounding steel capsule, the wave in the steel actually surrounds the powder capsule in a “pincer” action and penetrates along the lateral surface of the disc, as well as on the top surface. This specimen shows many cracks in the bottom region, where the microhardness values show the highest value. As reported earlier [5, 14] the microhardness distribution of shock-consolidated material shows the same tendency of temperature distribution during the passage of the shock wave. Higher hardness values are also obtained in the high shock temperature areas in our specimens. The highest microhardness value in Fig. 5b (46.0 GPa) is still low, compared with the value of single-crystal diamond (120 GPa [18]). This suggests that the bonding between particles is still imperfect in this sample. Fig. 5c shows the compacted 20–25 μm diamond powders (1-3). The enlarged scanning electron micrograph from the dark areas (see arrows in Fig. 5c) is shown in Fig. 5d. Small round diamond particles (0.5–1 μm) were observed in this sample (see arrows). Because the original diamond powder is monocrystalline, these small diamond particles are thought to be generated during the cooling of the molten surface of the particles. These equiaxed grains were also found in shock-synthesized diamond powders [19, 20]. The size of these small grains decreases with a decrease in the size of the original diamond particle, because of the variation in the cooling rate of the molten layer [21, 22]. Akashi and Sawaoka [5] reported that 2–20 μm was the optimum starting particle size for shock consolidation of diamond at room temperature. Potter and Ahrens [1] reported the same results. When the original particle size is very small ($<4\ \mu\text{m}$), the temperature of the powder surface is not sufficient to generate the molten layer to enhance the bonding. When the particle size is too large, many cracks are generated in the particles by the high pressure and a transformation from diamond to graphite is caused by the thick molten layer [1], which cools down at a slower rate.

3.2. Characterization of hot-consolidated diamond and graphite powder mixture

The consolidated diamond and graphite powder mixture (1-4) shows an extremely high microhardness value: $>55\ \text{GPa}$ (Table I). The present high hardness values confirm Potter and Ahrens' [2] observations which demonstrated the improvement of the bonding by the addition of graphite to diamond at room temperature. These authors suggested that the graphite, which has a lower thermal diffusivity, delays thermal equilibrium between the molten surface and the interior of the diamond. This heat enhances the



26	20	23	15	14	13
12	19	21	28	33	25
24	30	33	46	40	38

(b)

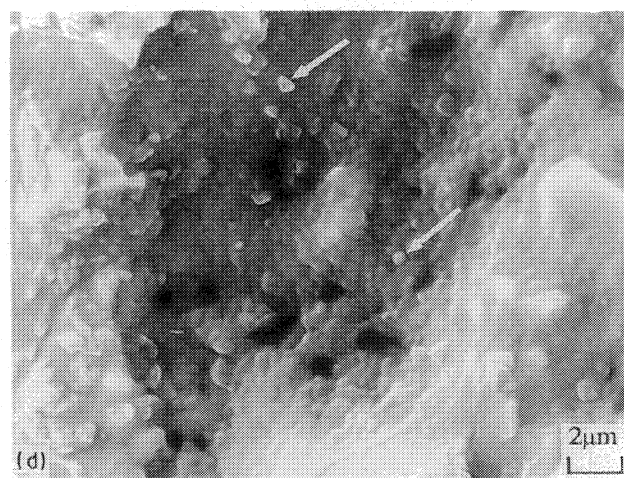
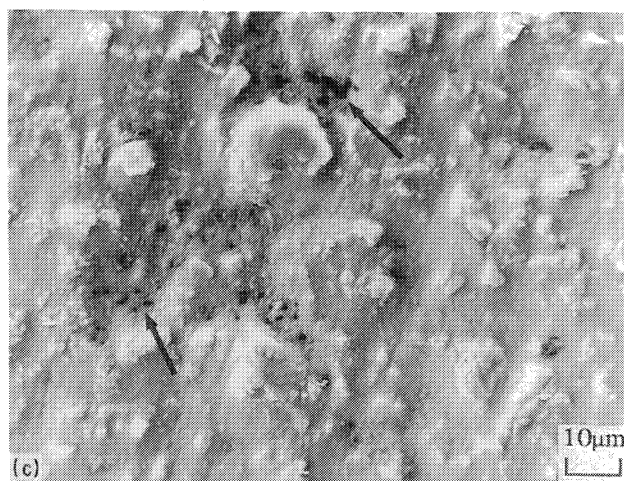


Figure 5 (a) Cross-section of consolidated 20–25 μm diamond (1-3), and (b) its Vicker's hardness distribution in GPa, (c) scanning electron micrograph from the higher hardness regions, (d) enlarged scanning electron micrograph from the dark areas.

bonding between particles. Deformation of the softer component graphite also decreases the pressure concentration and fills the pores between diamond particles. X-ray diffraction showed that part of the graphite powders was transformed to diamond. In the case of the second experiment (2-4), agglomeration of the graphite powders was observed in the sample. This diamond/graphite composite was strongly affected by the distribution of graphite powder. The agglomeration of graphite due to poor mixing may have contributed to the lower hardness of this specimen (25.8 GPa); the non-uniform distribution impeded its retention in the diamond form and its strengthening/bonding effect.

3.3. Characterization of hot-consolidated diamond and silicon powder mixture

The recovered samples of compacted diamond and silicon powders (1-5, 2-5) indicated a poor consolidated appearance with considerable cracking. Micro-Vicker's hardness tests were conducted and resulted in a hardness of about 7.4–15.8 GPa. Degradation of consolidated diamond powders appeared with the addition of silicon powders. Scanning electron micrographs and silicon X-ray mapping (Fig. 6)

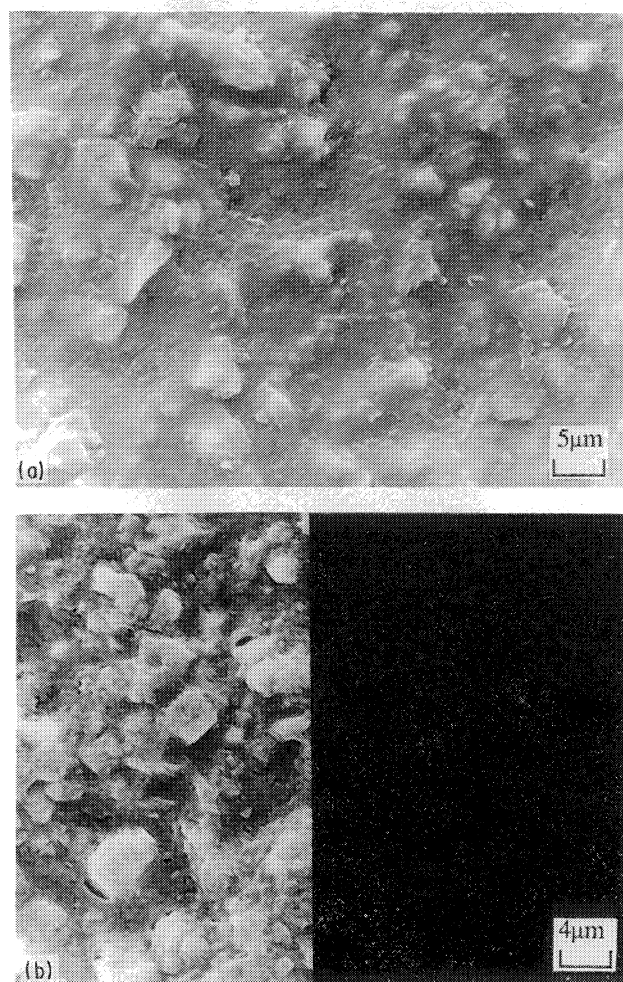


Figure 6 Scanning electron micrographs and silicon X-ray mapping of diamond/silicon composites.

show that the silicon powders are very well dispersed in the diamond powders. However, we expected the heat generated from the reaction between silicon and carbon to enhance the bonding, as suggested by Potter and Ahrens [2] and others [13–15] between particles, but in this case the results are considerably less successful than the diamond with graphite experiments.

3.4. Characterization of hot-consolidated c-BN powders

The recovered c-BN samples are very well consolidated. SEM images are shown in Fig. 7a and b which demonstrate good bonding between the c-BN powders. Fig. 7a is a fracture surface of 10–20 μm c-BN (1-6) which suggests the melting of particle surfaces. In some areas of this specimen, small voids are observed, probably due to the gas generated by the decomposition of c-BN (Fig. 7b). In comparison with the first experiment, sample 2-6 (second experiment) shows very high hardness (61.4 GPa) and good bonding (Fig. 8) which are due to the higher preheating and impact velocity. In the first experiment, a very well consolidated 40–50 μm c-BN sample (1-7) showing high microhardness value (53.0 GPa, Table I) was

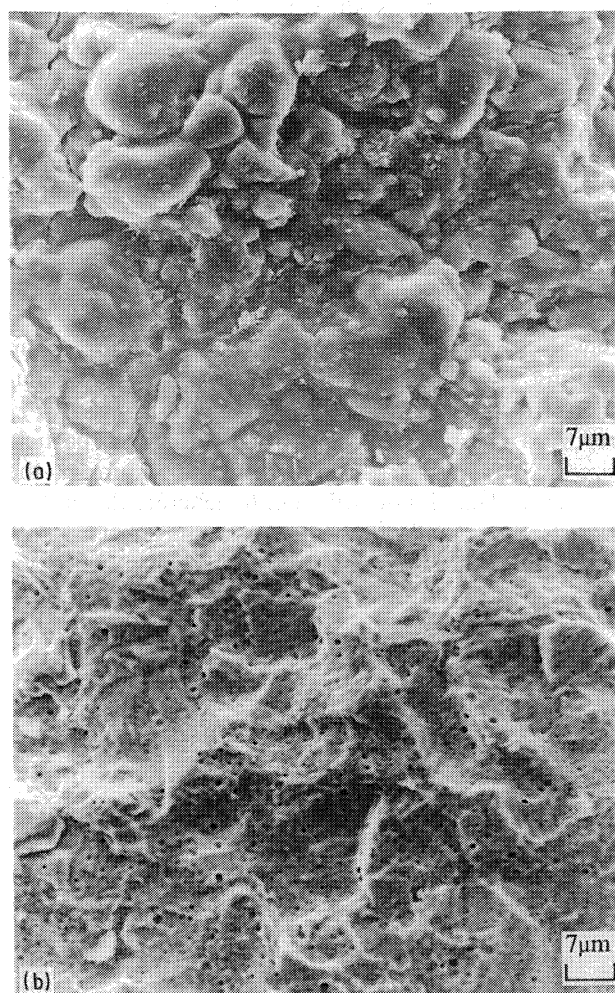


Figure 7 (a) Scanning electron micrographs of fracture surface of consolidated 10–20 μm c-BN (1-6); (b) small voids generated in this sample.

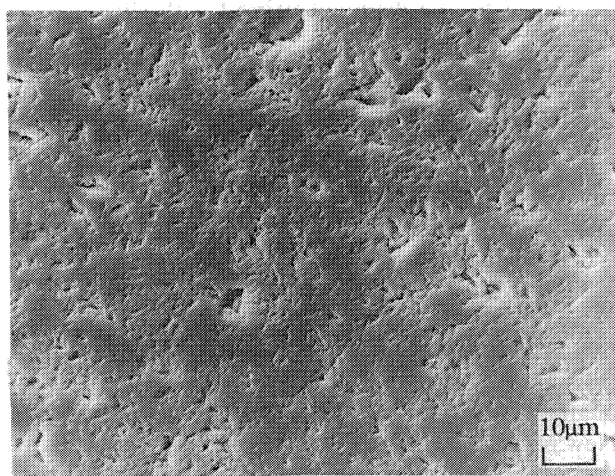


Figure 8 Scanning electron micrograph of polished surface of consolidated 10–20 μm c-BN powders (2-6).

also obtained which is comparable to the c-BN (60–75 GPa) synthesized by static high pressure and temperature [18]. A fractograph of the specimen is shown in Fig. 9a; the crack propagates through the particles, which indicates that the bonding of the powders is excellent. However, in the case of the second experiment (2-7), small amounts of the graphite phase of BN were detected by X-ray diffraction due to a higher preheat and impact velocity. This phase transformation weakens the bonding between c-BN powders (Fig. 9b) and causes degradation of consolidated c-BN samples. The microhardness values were relatively lower, near 29.9 GPa. Comparison of the X-ray peaks (Fig. 10) for the compacts 2-6 and 2-7 shows phase transformation in the BN powder after hot shock consolidation. As Akashi and Sawaoka [6] reported for the shock consolidated c-BN powders at room temperature, the specimen with larger particle size showed higher hardness value. However, due to the phase transformation that occurred within c-BN powders, this is a disadvantage for hot shock consolidation.

3.5. Characterization of hot-consolidated c-BN and graphite powder mixture

SEM observation of the recovered samples (1-8) showed a weak bonding between c-BN and graphite powders. The graphite powders are severely deformed and uniformly distributed on the surfaces of the c-BN powders after shock consolidation. The consolidated c-BN admixed with graphite shows low microhardness value, compared with the c-BN without graphite (Table I). It is considered that the cooling rate was not sufficiently high for the transformation of the graphite into diamond. The remaining graphite causes the degradation of consolidated samples.

3.6. Characterization of hot-consolidated c-BN and titanium plus carbon powder mixture

Elemental powders (titanium and graphite) were added to c-BN powders (2-9). The basic idea was to

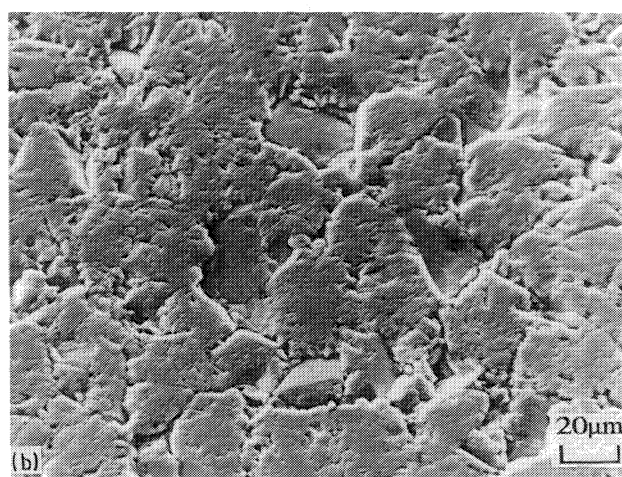
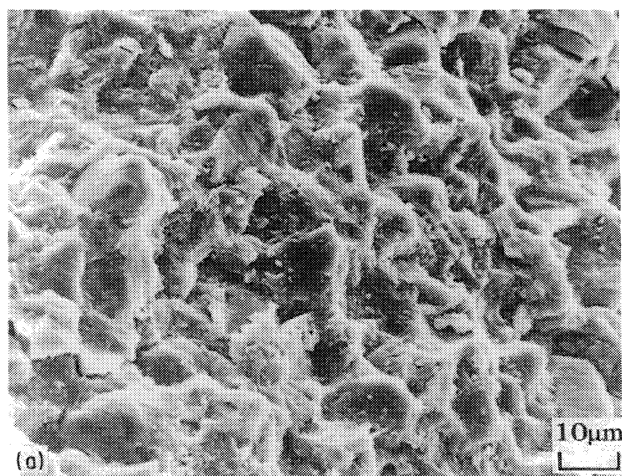


Figure 9 (a) Scanning electron micrograph of fracture surface of 40–50 μm c-BN (1-7) showing a crack propagating through the particles, and (b) polished surface of 40–50 μm c-BN (2-7) showing that the bonding of the powder is weaker.

use the preheat and passage of the shock wave to activate highly exothermic reactions ($\text{Ti} + \text{C}$) in order to provide enhanced bonding between the c-BN powders. However, SEM images (Fig. 11) showed that the reaction did not take place; the microhardness was about 16.3 GPa. In this case, the titanium and carbon powders were deformed more intensely than the c-BN particles (because of the lower yield stress of titanium and carbon) and they just interlocked together.

4. Conclusions

Diamond and cubic boron nitride powders were shock consolidated at high temperatures (873 and 973 K) by using a planar impact system. Hot shock consolidated materials exhibited a decrease of surface cracks compared with specimens consolidated at room temperature. The consolidated diamond specimen showed evidence of surface melting of the particles by small crystallized grains generated during cooling of this molten layer. The consolidated 4–8 μm diamond powders showed good bonding. It is reported that c-BN is more stable than diamond at high temperatures in an oxygen atmosphere [18]. This

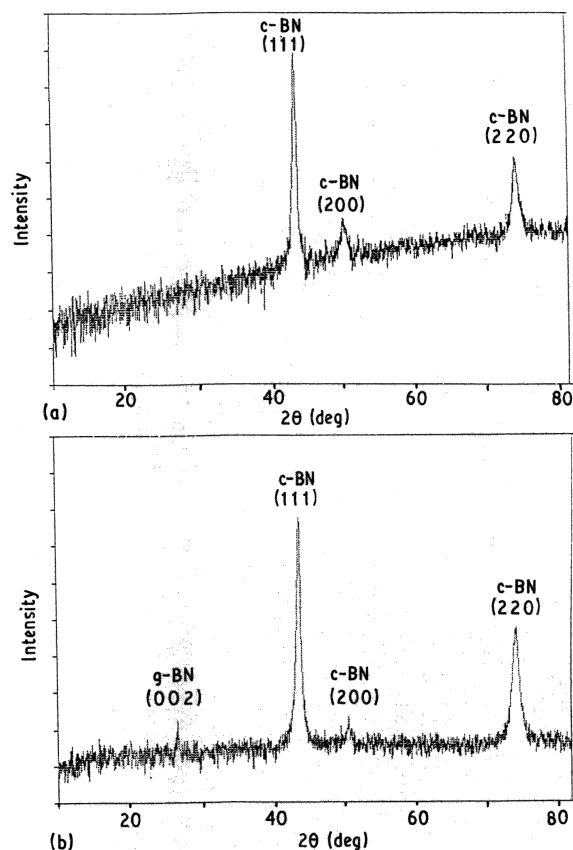


Figure 10 X-ray analysis results of c-BN compacts (a) hot-shocked sample (2-6), (b) hot-shocked sample (2-7) showing the generation of the graphite phase of BN.

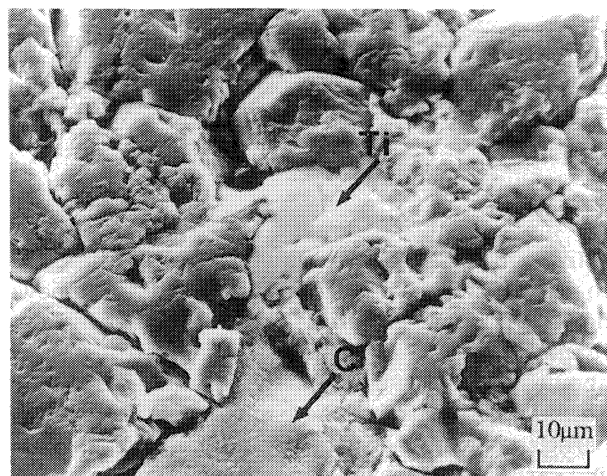


Figure 11 Scanning electron micrograph showing that reaction (Ti + C) did not take place in the compacted c-BN powders.

suggests that the transformation from cubic to hexagonal boron nitride rarely occurs even when the cooling rate is slow, compared with the diamond powders. Excess cracks and generation of graphite were observed when the particle size was increased. Hot consolidated diamond admixed with graphite showed the highest Vickers's microhardness value (>55 GPa); the improvement in the bonding was not

confirmed by the diamond mixed with silicon powders. Well-consolidated c-BN samples were obtained and showed a very high hardness value (>53 GPa). The improvement in the bonding of c-BN powders by the addition of graphite or titanium plus carbon could not be recognized in the present investigation.

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