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Effect of c-BN Size and Content on the Self-Propagating High-Temperature Synthesis of c-BN Composites Bonded with Ti-Al-C System Multiphase Products

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Abstract: A mixture of Ti, Al, graphite and c-BN powders was used as raw material to fabricate Ti_2AlC matrix-bonded c-BN composite using the self-propagating high-temperature synthesis (SHS) method. The effect of c-BN size and content on the fabrication of the composites was investigated. The results show that Ti_2AlC matrix-bonded c-BN composites can be obtained by SHS. c-BN content and size evidently affected the phase composition and microstructure characteristic of the composites. At 10–30% c-BN (120/140 mesh) content, the product phases were Ti_2AlC, Ti_3AlC_2, Al_3Ti, TiN, TiC, AlN, graphite and TiB_2. A dense transition layer with a thickness of about 10 μm showed the interface between c-BN and the matrix. However, Al peaks appeared, and the titanium aluminium carbide peak became weak in the samples containing 40% and 50% c-BN. c-BN was unequally enwrapped by one coating with a thickness of about 2 μm. The main product phases of the samples were Ti_2AlC, Ti_3AlC_2, Al_3Ti, TiN, TiC, AlN, graphite and TiB_2 in the products with different c-BN sizes. The addition of coarse c-BN particles (80/100, 120/140 and 170/200 mesh) yielded a transition layer with a thickness of approximately 10 μm on the interface and Ti_2AlC main phase matrix. The finer the particle size, the greater the reaction activity. When c-BN was finer (20 μm), c-BN more easily reacted with Ti and Al to form TiN, AlN and TiB_2. The synthesis of Ti_2AlC was obviously inhibited.

Key words: titanium aluminium carbide, c-BN, content, size, self-propagating high-temperature synthesis

Introduction

Ceramic-bonded c-BN tools possess high strength, high grinding efficiency and low thermal expansion coefficient [1,2]. Ceramic-bonded c-BN wheels, as one of the rapidly developed superhard grinding materials, have been applied in various engineering applications including in the machining of cemented carbide, metal ceramics, ferrite and sapphire. The self-propagating high-temperature synthesis (SHS) method is interesting because of its advantages such as very short processing time, low energy requirement and high-purity products [3,4]. Diamond tools have been fabricated by SHS method [5–9]. The SHS systems include Ti-Al-C-diamond, Ti-B-diamond, Ni-Al-Cu-Ti-diamond and Ni-Al-diamond. However, few studies have reported on the SHS fabrication of c-BN abrasive tools.

MAX phase ceramics, such as Ti_2AlC, have both metallic and ceramic properties [10,11]. These materials are relatively ductile and machinable. They also possess low density, good electrical and thermal conductivities as well as excellent anti-oxidation and thermal shock resistance. Thus, MAX phase ceramics have good potential as carrier-bonded materials for c-BN. We fabricated Ti_2AlC- and Ti_3AlC_2-bonded polycrystalline c-BN tools by SHS technology in our previous study [12]. This result shows that the diamond bonds well with Ti_2AlC or Ti_3AlC_2 matrix.

In this paper, we report the fabrication of a c-BN material bonded with Ti-Al-C system multiphase products using the SHS method. The effect of c-BN size and content on the fabrication process was investigated.

Experimental procedure

Powders of Ti (99.5% pure, ~53 μm), Al (99.6% pure, ~53 μm) and graphite (99% pure, ~53 μm) were mixed in a mole ratio calculated from 2Ti/Al/C. Then, c-BN powders (W20; 170/200, 120/140 and 80/100 mesh) were added in the above mixed powders. These powders

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were ball-milled in a planetary ball mill (XM-4 × 05) for 5 h using agate balls to achieve homogeneous mixed mixtures. The rotation speed was set to 50 rpm to avoid alterations in the c-BN size. Finally, the mixtures were cold-pressed into 4 mm × Φ13 mm bars. The samples were mounted in a container, in which argon was continuously supplied to provide an inert environment. The combustion reactions were conducted in a CO_2 laser. The laser was operated at an output power of 2 kW and beam diameter of 8 mm. The temperature was measured using a high-temperature infrared radiation thermometer (Model EC8300). The X-ray diffraction (XRD) experiments were performed in a rotating anode X-ray diffractometer (Rigaku Ultima IV) with Cu Kα radiation. Scanning electron microscopy was conducted using a VEGA3 TESCAN microscope equipped with an energy-dispersive spectroscope (EDS).

Results and discussion

Figure 1 shows the typical temperature profile recorded from the combustion of the Ti/Al/C/c-BN sample. The maximum combustion temperature reached 1,977°C. The high combustion temperature evidently enhanced the synthesis of Ti-Al-C system multiphase products.

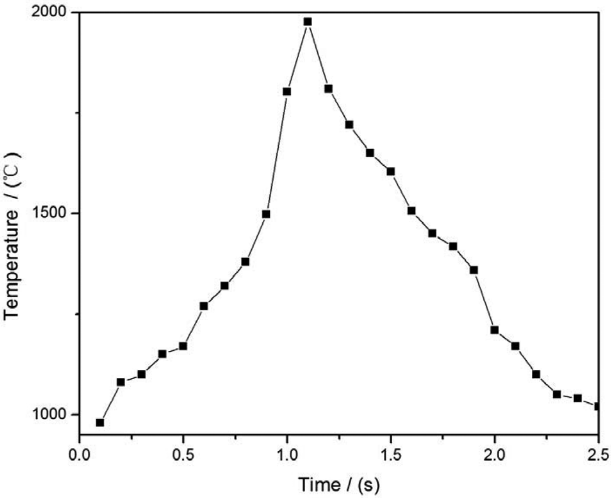

Figure 1: A typical temperature profile recorded from the combustion of the Ti/Al/C/c-BN reactants.

The XRD patterns of the samples with different c-BN (120/140 mesh) contents obtained by SHS are shown in Figure 2. The main product phases of the sample were Ti_2AlC, Ti_3AlC_2, Al_3Ti, TiN, TiC, AlN, graphite and TiB_2.

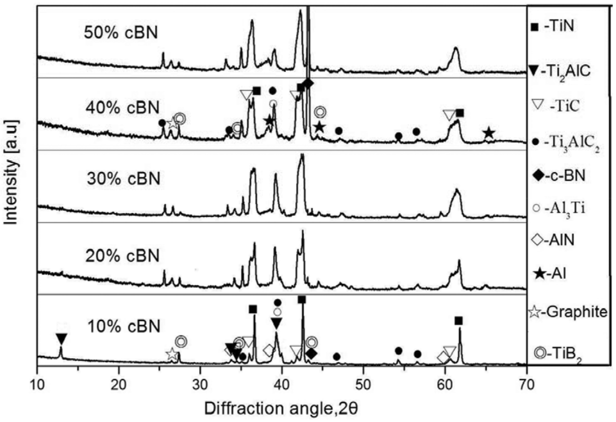

Figure 2: The XRD patterns of the samples that contained different c-BN contents obtained by SHS.

c-BN evidently participated in the reaction of the Ti-Al-C system and formed some related compounds. The possible reactions were as follows:

$$3Ti + Al + 2C \rightarrow Ti_3AlC_2 \tag{1}$$

$$2Ti + Al + C \rightarrow Ti_2AlC \tag{2}$$

$$Ti + 2Al + 2BN \rightarrow TiB_2 + 2AlN \tag{3}$$

$$2Ti + 2BN \rightarrow TiB_2 + TiN \tag{4}$$

$$Ti + C \rightarrow TiC \tag{5}$$

$$Ti + 3Al \rightarrow Al_3Ti \tag{6}$$

The c-BN peaks were relatively weaker at low c-BN content (10%, 20% and 30%). However, the c-BN peaks became very strong when the c-BN content reached 40% and 50%. Additionally, Al peaks appeared and the titanium aluminium carbide peak became weak in the samples containing 40% and 50% c-BN. These results show that c-BN sufficiently reacted with the matrix element when its content was lower than 40% in the raw materials. Moreover, the addition of excess c-BN may inhibit the synthesis of titanium aluminium carbide and lead to insufficient reaction of the raw materials.

These results indicate that c-BN reacted with Ti and Al to some extent. The addition of different amounts of c-BN obviously affected the reaction process and result of the Ti-Al-C system.

Figure 3 shows a fracture micrograph of the synthesized sample containing 20% c-BN (120/140 mesh). A dense transition layer with a thickness of approximately 10 μm presented the interface between c-BN and the matrix (Figure 3(a)). The transition layer was composed of TiN,

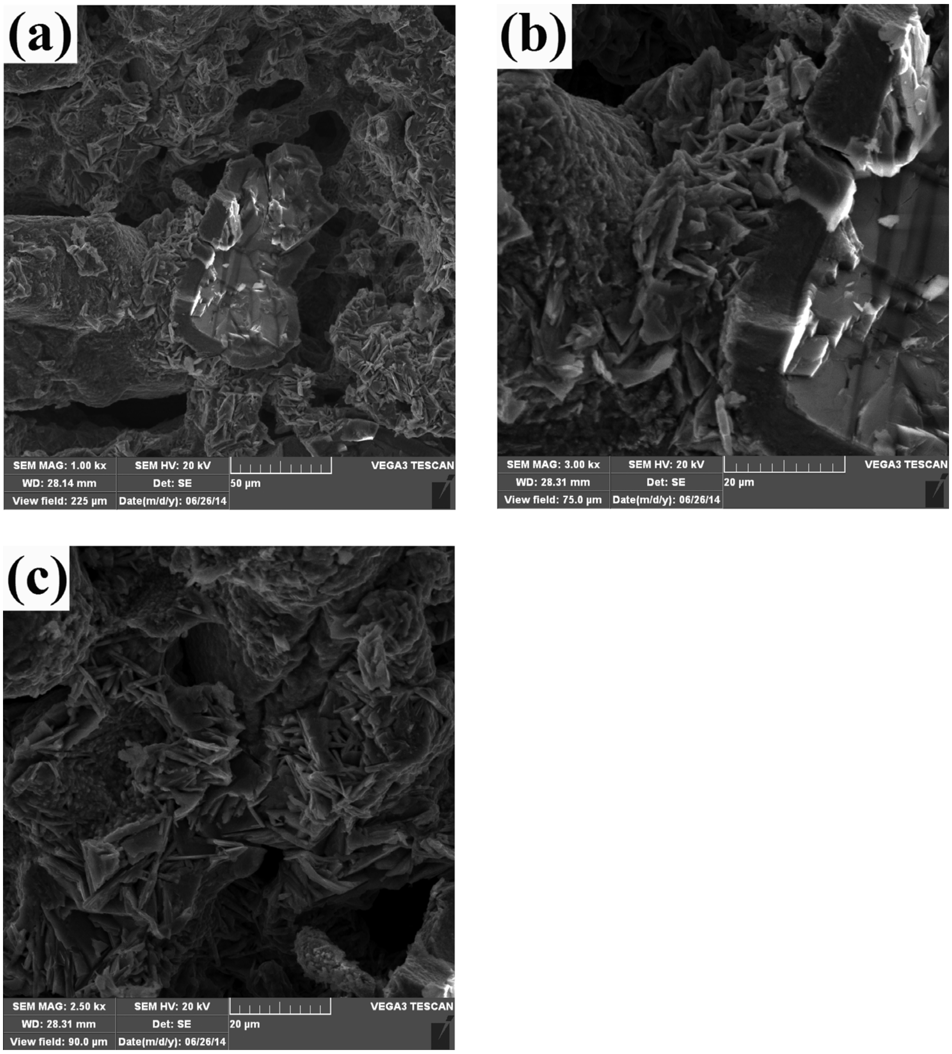

Figure 3: Fracture micrograph of the synthesized sample containing 20% c-BN (120/140 mesh). (a) low latitude morphology, (b) the interface morphology between c-BN and the matrix, (c) the morphology of matrix.

AlN and TiB_2, which is in accordance with the XRD result. The interface between c-BN and the matrix was well-knit, indicating that c-BN bonds well with the matrix in this sample (Figure 3(b)). The matrix (Figure 3(c)) was basically composed of lamellar Ti_2AlC or Ti_3AlC_2 grains and particle TiC grains, which were identified by EDS. The Ti_2AlC or Ti_3AlC_2 grains had approximate lengths of 10 μm to 20 μm. TiC grains had an average size of approximately 2 μm. Figure 4 shows a fracture micrograph of the synthesized sample that contained 0% c-BN. The c-BN was unequally enwrapped by one coating (Figure 4(a)), and many particles with varied sizes were dispersed in the coating (Figure 4(b)). The coating had a thickness of approximately 2 μm. The XRD and EDS results show that the coating was composed of Al, TiN, TiB_2 and AlN. Additionally, the matrix was composed of some titanium aluminium carbide and amounts of particles (Figure 4(b)). Notably, the Ti_2AlC or Ti_3AlC_2 grains became obviously fine.

Thus, the c-BN additive amount obviously affected the reaction of Ti-Al-graphite-c-BN systems. The primary reasons are as follows.

The SHS adiabatic temperature of 2Ti/Al/C system was approximately 1,600–1,800°C [11]. The transform temperature of c-BN to h-BN was usually 1,400°C. Some amounts of c-BN may transform to h-BN because of the extremely short reaction time (2–3 s) of SHS. Therefore, the B and N elements on the surface of c-BN reacted with Ti and Al to form the TiB_2, AlN and TiN phases, which ensured a good bond between c-BN and Ti_2AlC matrix. However, this phenomenon led to the shortage of Al and Ti, which reacted with graphite. Some graphite cannot react, and residue was found in the products. Hence, the composition of the raw materials deviated from the stoichiometric ratio of Ti_2AlC phase to some extent. Finally, the Ti_2AlC peaks in the bonded samples were slightly weaker than that of the samples without c-BN [11].

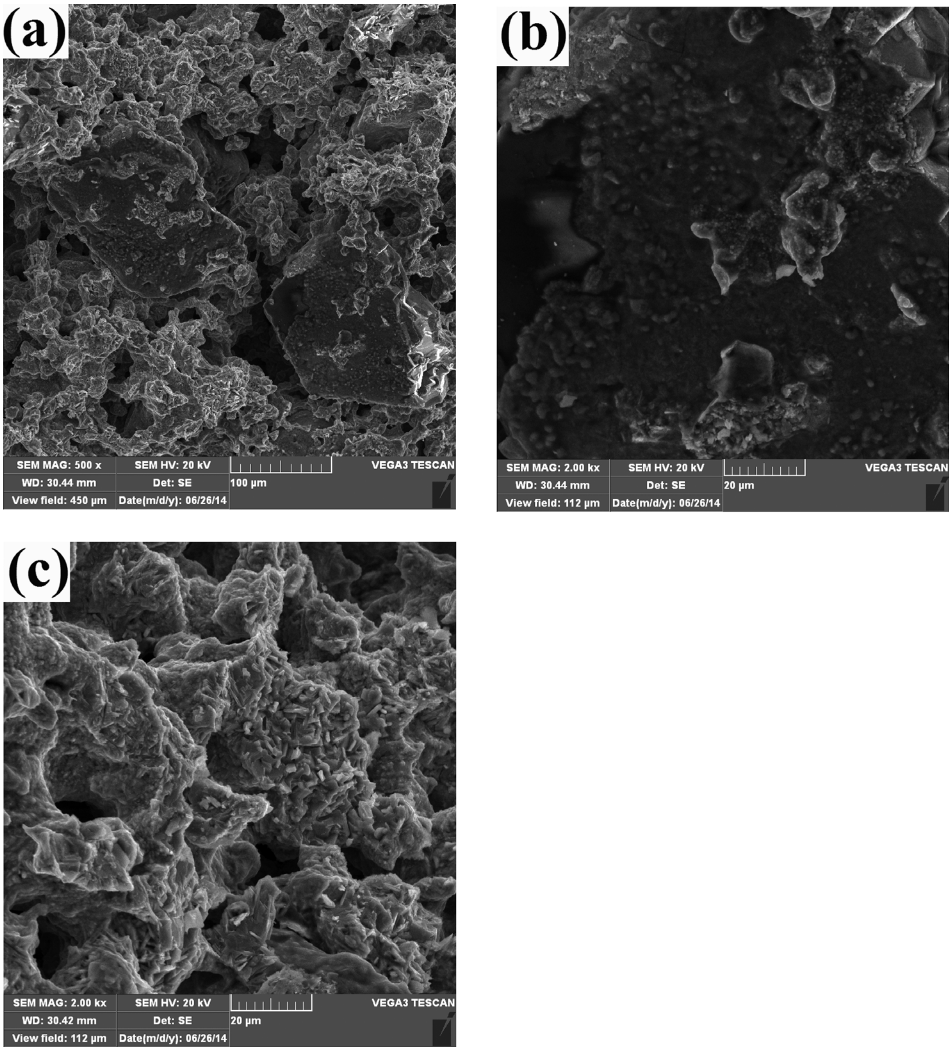

Figure 4: Fracture micrograph of the synthesized sample containing 50% c-BN (120/140 mesh).
(a) low latitude morphology,
(b) the surface morphology of c-BN,
(c) the morphology of matrix.

However, at 40% and 50% c-BN content, the good heat conduction of c-BN played a key role, that is, it apparently decreased the adiabatic temperature of the system. When c-BN content was lower than 40%, c-BN reacted adequately with the Ti and Al. The reaction degree was reduced with increasing c-BN content. Therefore, complete c-BN particles were observed in the sample that contained 50% c-BN (Figure 4(a)). Excessive c-BN addition (40% or higher) seriously decreased the adiabatic temperature, which inhibited the reaction synthesis of Ti-Al-C system. This phenomenon caused the incomplete reaction of the raw materials. Consequently, Ti_2AlC peaks obviously became weak, and Al peaks appeared in the samples that contained 40% and 50% c-BN.

Hence, addition of different c-BN contents affected not only the reaction characteristic of the Ti-Al-C system, but also the bonding condition between c-BN and the matrix. Considering the good bond of c-BN with the matrix in the sample that contained 10% c-BN, 10% c-BN addition was used in the following experiment.

Figure 5 illustrates the XRD patterns of the samples containing 10% c-BN with different particle sizes obtained by SHS. The product phase of the sample was Ti_2AlC, Ti_3AlC_2, Al_3Ti, TiN, TiC, AlN, graphite and TiB_2 in the products. Ti_2AlC and Ti_3AlC_2 peaks in the samples containing coarser c-BN were much stronger, which indicates that Ti_2AlC and Ti_3AlC_2 contents were higher in the samples containing coarser c-BN. However, when 20 μm c-BN was added, Al peak appeared, and the titanium aluminium carbide peaks vanished. The main phases of this sample were TiC, TiN, AlN and TiB_2. These results show that the c-BN size obviously affects the phase composition of the matrix. Finer c-BN size may inhibit the synthesis of Ti_2AlC.

Figure 6 shows a fracture micrograph of the synthesized sample containing 20 μm c-BN. The sample was

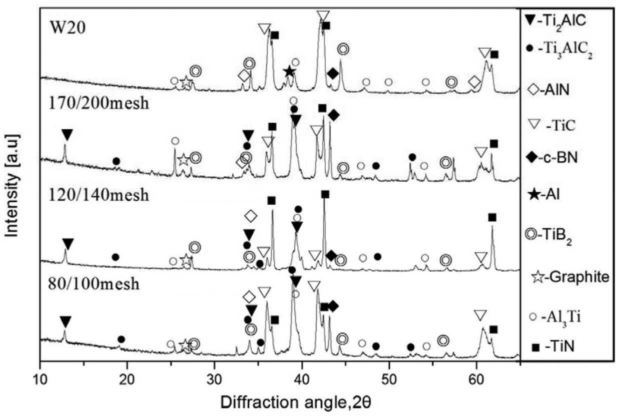

Figure 5: The XRD patterns of the samples that contained different c-BN (10%) particle sizes obtained by SHS.

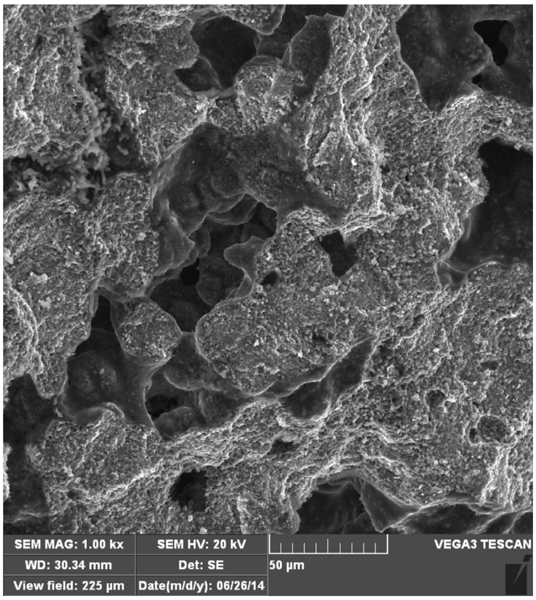

Figure 6: Fracture micrograph of the synthesized sample that contained 20 μm c-BN.

basically composed of granules. The c-BN particles were difficult to find in its microstructure. Figure 7 shows a fracture micrograph of the sample containing 20 μm c-BN. The c-BN particles were closely inserted in the matrix (Figure 7). A transition layer with a thickness of 2–5 μm was formed on the surface of c-BN (Figure 6). The layer had a good bond with c-BN and the matrix. Ti_2AlC and Ti_3AlC_2 lathed grains were found near the c-BN. Figure 8 shows a fracture micrograph of the sample containing 125 μm c-BN, which shows a morphology that was very similar to that of the 120/140 mesh c-BN (Figure 3). c-BN also bonded well with the matrix.

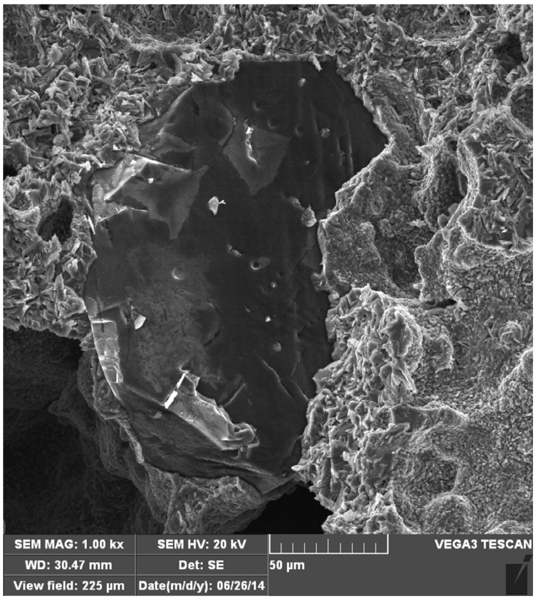

Figure 7: Fracture micrograph of the synthesized sample containing 170/200 mesh c-BN.

Thus, the addition of different c-BN particle sizes apparently affects the SHS process. Under high-temperature SHS reaction, c-BN with fine particle size can fully transform to h-BN. Therefore, finer c-BN particles more adequately participate in the reaction with the matrix element. Consequently, the addition of coarse c-BN particle can yield the Ti_2AlC main phase matrix, and c-BN can adequately react with Ti and Al to form TiN, AlN and TiB_2 accordingly. Obviously, fine c-BN was not suitable as the additive for these composites.

Conclusions

c-BN materials bonded with Ti-Al-C system multiphase products can be obtained by SHS method. The main product phases were Ti_2AlC, Ti_3AlC_2, Al_3Ti, TiN, TiC, AlN, graphite and TiB_2. The addition of various c-BN particle sizes and contents evidently affects the SHS process between c-BN and matrix. The following conclusions could be drawn:

(1) At 10–30% c-BN (120/140 mesh) content, the matrix was mainly composed of Ti_2AlC, Ti_3AlC_2, Al_3Ti and TiC. A dense transition layer with a thickness of approximately 10 μm appeared as the interface of c-BN in these samples. However, Al appeared and titanium aluminium carbide content decreased in the samples containing 40% and 50% c-BN. c-BN

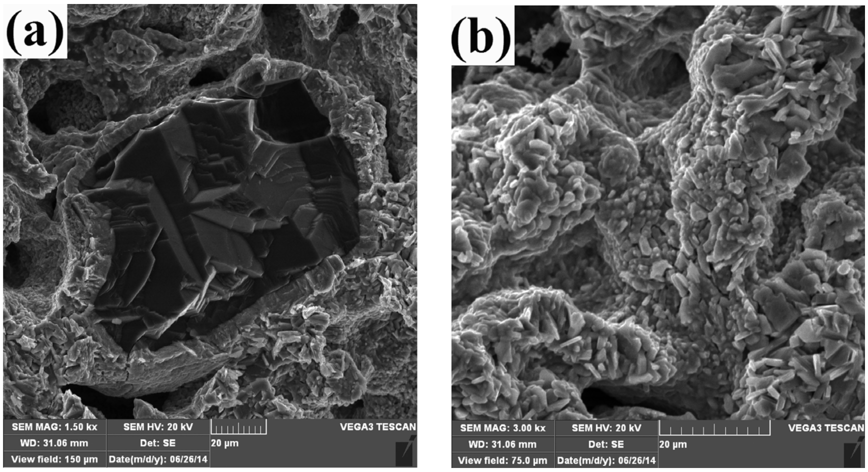

Figure 8: Fracture micrograph of the synthesized sample containing 80/100 mesh c-BN the morphology of (a) c-BN and (b) the matrix.

was unequally enwrapped by the coating with a thickness of approximately 2 μm.

(2) When 20 μm c-BN was added, c-BN adequately reacted with Ti and Al to form TiN, AlN and TiB_2. The synthesis of titanium aluminium carbide was inhibited. Addition of coarser c-BN particles (80/100, 120/140 and 170/200 mesh) yielded a dense transition layer in the samples. Additionally, the main phase matrix of titanium aluminium carbide was also obtained.

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