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cBN BASED MATERIALS WITH TiN-Al BINDER PHASE: SINTERING, STRUCTURE, PROPERTIES

Cubic boron nitride material was produced via high pressure and high temperature sintering with titanium nitride and aluminum as binder phase. Microstructure, phase composition and physical-mechanical properties of the material were subject of current research. Formation of TiB_2 and AlN was established in samples sintered at temperatures above 1750 °C. The highest values of microhardness were found in samples sintered at the temperature range of 1850 – 2000 °C. Fracture toughness of samples practically does not change during thermobaric sintering.

Key words: *high pressure apparatus, superhard ceramics, cBN, phase composition, structure, titanium*

Introduction

Cubic boron nitride (cBN) based materials are well known for their resistance to chemical wear and excellent physical and mechanical properties [1]. Sintering of pure cBN is complicated due to its covalent bounding. Therefore, different types of additives or binder phases are used to promote sintering process and improve mechanical properties. The most common type of binder phase are the elements from groups IV–VI of the Periodic table and their refractory compounds [2–6]. Pure aluminum, cobalt, zirconium and nickel also can be used as a binder phase [7, 8]. Reaction between cBN and Al under a relatively low temperatures is provided by the low melting temperature of Al at ambient pressures [9]. The final microstructure and phase composition are the major determination of the properties of the cBN-based materials.

Comparison of theoretical calculations and experimental results in cBN–TiN and cBN–TiC systems (in molar ratios 1:1 and 2:1), conducted by E. Benko confirmed formation of TiB_2 in cBN-TiN system and TiB_2 with $TiC_{0,8}N_{0,2}$ in cBN-TiC system. It was assumed that reducing the amount of titanium nitride and especially titanium carbide though formation of borides and nitrides in the process of heat treatment ($p = 3 \cdot 10^{-3}$ Pa, $T = 1000$ °C, $T = 1400$ °C, $\tau = 1$ h) leads to increasing of microhardness [10,11]. It was found that in condition of HPHT sintering of cBN–TiN $p = 7{,}5$ GPa, $T = 1400 – 2000$ °C), increasing of cBN content from 38 vol.% to 65 vol.% has positive evidence on Young's modulus and microhardness [12]. An improvement of performance of the cBN–TiN materials

was achieved by addition of a small amount of Al [13]. Spark plasma sintering of materials in cBN–Ti–Al system leads to formation of TiN, TiB_2, and AlN, as a result of interaction between Ti and Al with cBN. Newly formed phases act as additional binder phases, which additionally increase mechanical properties, as well as increasing of sintering temperature up to 1700 °C [14]. cBN-based materials with titanium binders are widely studied and available on the market. Meanwhile improvement of composition and mechanical properties remains relevant from both scientific and commercial points of view.

Experimental procedure

The starting materials used for this work were crushed cBN powders (Element Six), TiN (ABRC) and aluminum flakes (ABRC). HPHT sintering was conducted in a toroidal high-pressure apparatus HPA-TOR 30 under a pressure of 7,7 GPa at temperatures in the range of 1750–2450 °C. Analysis of phase composition of the sintered samples was held using a STOE STADI MP X-ray diffractometer (XRD) in Cu-Kα radiation. Prior to microstructural and indentation studies, the samples were polished with diamond suspension using a Struers Tegramin. In order to obtain mirrorlike surface final polishing of samples was provided using metallographic vibratory polisher Giga-1200 with silica colloidal solution (0.04μm). Scanning electron microscope (SEM) LEO 1560 was used for analysis of the microstructures. Density of samples was measured by direct method. Speed of longitudinal and transverse velocity was estimated with using of Olympus 38D Plus for further calculation of elastic properties of samples. Microhardness and fracture toughness were determined using a THV-30MDX Vickers hardness tester. Leica light optical microscope was used for measurements of indenter imprints.

Results and discussion

Phase analysis of the initial powder mixture for sintering (T = 20 °C) confirmed presence of cBN (space group *F-43m* with lattice constants $a = b = c = 3.6162$ Å), TiN (*Fm3m*, $a = b = c = 4.2397$ Å) and Al (*Fm3m*, $a = b = c = 4.0486$ Å). Application of HPHT conditions promoted interaction between components of the mixture and phase composition of samples sintered at temperatures above 1750 °C complemented by titanium diboride TiB_2 (*P6/mmm*, $a = b = 3.0386$ Å, $c = 3.2311$ Å) and aluminium nitride AlN (*P63mc*, $a = b = 3.1100$ Å, $c = 4.9750$ Å) (Fig. 1). Macroscopic cracking of the sample sintered at 1750 °C occurred during the process of unloading of HPHT apparatus after sintering. That is why this sample was investigated only by using XRD analysis.

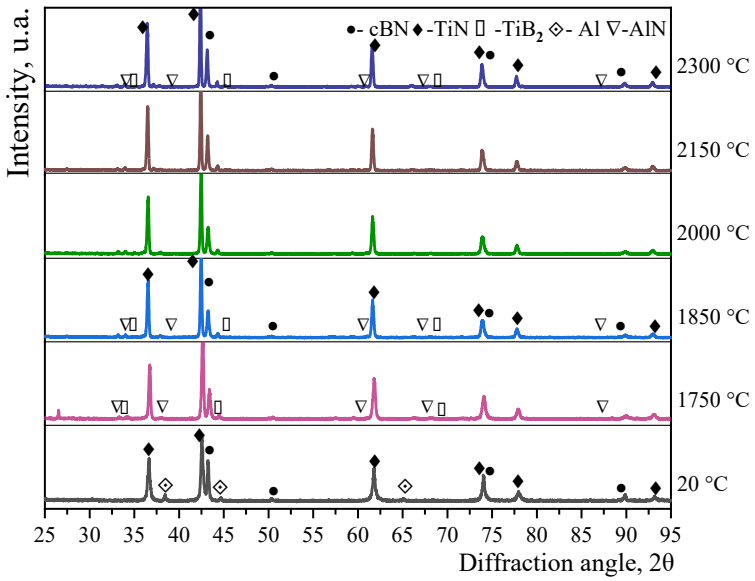


Fig. 1. X-Ray diffraction pattern of initial mixture for sintering (20 °C) and sintered samples in system cBN–TiN–Al, p = 7,7 GPa

On high resolution images (Mag. 60000^x) of the microstructures of samples sintered at the temperature range of 1850–2300 °C the formation of reaction products are clearly visible, especially in the sample sintered at 2300 °C, where completeness of the chemical reaction is the highest (Fig. 2).

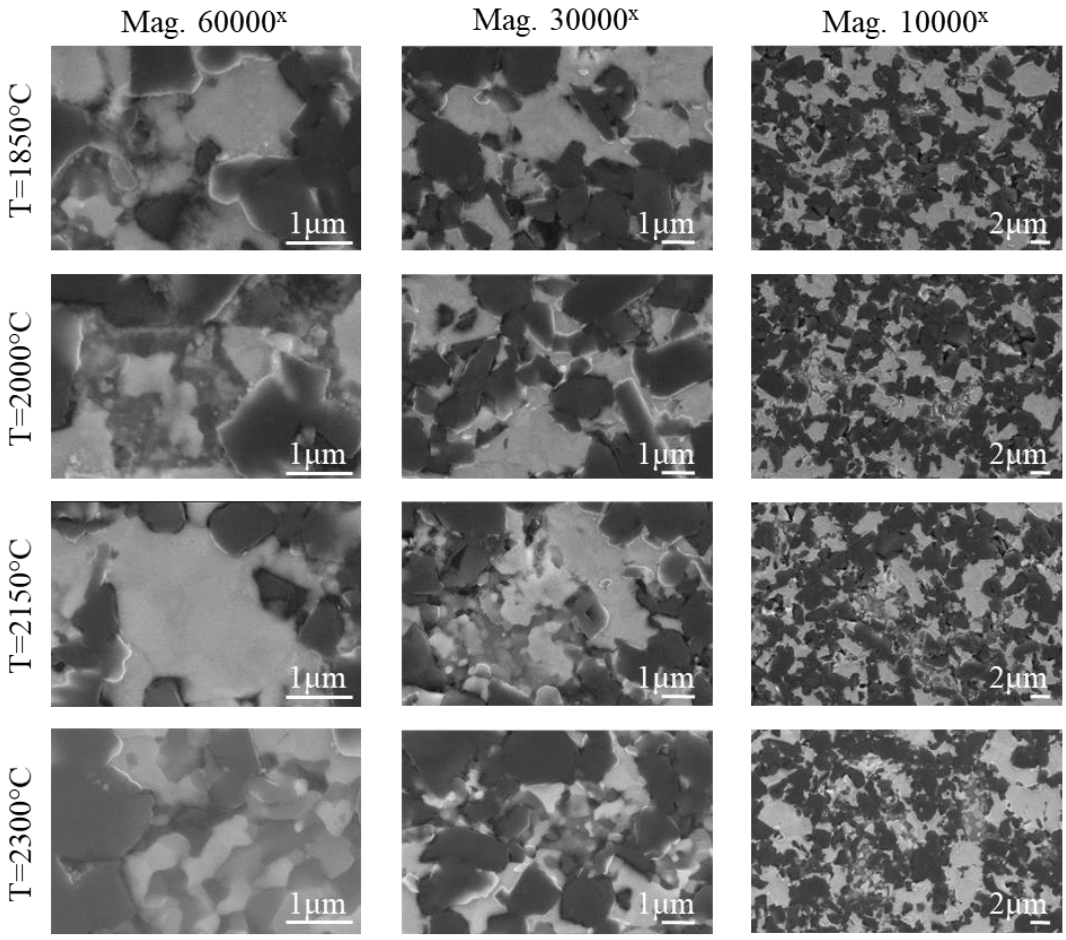


Fig. 2. SEM images of sintered samples from cBN–TiN–Al system

The reaction products can be observed in the intergrain space between the cBN particles, where the original phases of cBN, TiN and Al had been in contact to each other. Images obtained at lower magnification demonstrated that structures are homogeneous and no binder phase agglomerations larger than 4 μm were found in samples sintered at temperature range of 1850–2150 °C. It can be assumed that in samples sintered at 2300 °C a collective recrystallization occurs (Fig. 2).

The density of the green compact under the pressure of 7,7 GPa at temperature 20 °C – 3,2 g/cm^3. The density of the samples essentially increased at temperatures above 1850 °C and reached values above ≈ 4,08 g/cm^3. Further increasing of the sintering temperature showed limited effect on the densification of the compacts and the density dependence has the form of an asymptotic approximation to a certain limit corresponding to the densities of the initial powders and reaction products (Fig. 3). Young's and Shear modulus have similar dependence on sintering temperature (Fig. 4). Firstly, both of parameters slightly decrease at 2000 °C. This can be related to early step in formation of new phases and contamination of the grain boundaries. Secondary, at sintering temperatures above 2150 °C both modulus increase. This correlates with the increasing of amount of high modulus compound TiB_2. Microhardness of sintered samples also increased with application of

sintering temperature and amount of TiB_2, while fracture toughness of samples stayed in the range of mistake during all experiments. Decreasing of microhardness at 2300 °C occurred due to recrystallization process.

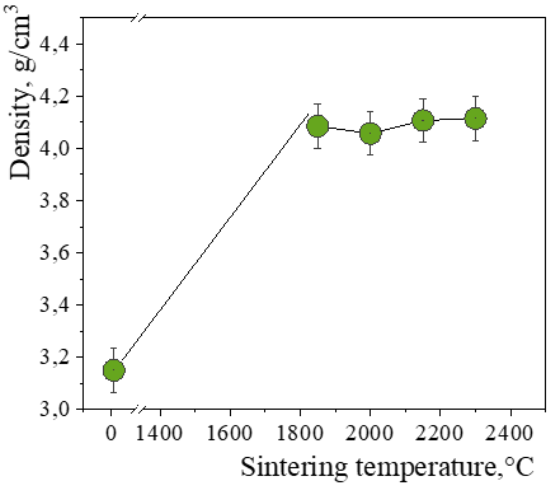
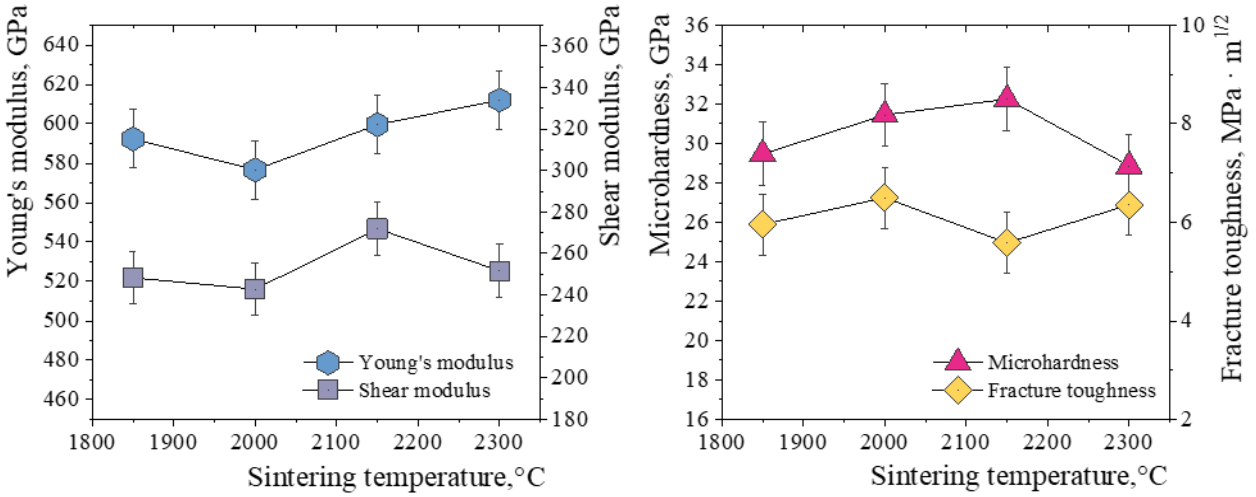


Fig. 3. Influence of sintering temperature on density of sintered samples



a *b*

Fig. 4. Influence of sintering temperature on a – physical and b – mechanical properties of sintered samples

Conclusions

It can be concluded that during HPHT sintering of cBN-based materials with Ti-Al binder the highest level of mechanical properties was achieved in the temperature range of 2000 – 2150 °C. Classical interaction between components of the mixture leads to formation of TiB_2 and Al at temperatures above 1750 °C. The reaction products took place in the intergrain space between the cBN particles.

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Методом спекания при высоком давлении и высокой температуре изготовлен материал на основе кубического нитрида бора с использованием нитрида титана и алюминия в качестве

связующей фазы. Объект исследования – структура, фазовый состав и физико-механические свойства материала. Установлено, что при температурах спекания выше 1750 °С в результате взаимодействия между компонентами шихты для спекания происходит образование диборида титана TiB_2 и AlN. Наивысшие значения микротвердости демонстрируют образцы, изготовленные в температурном интервале 1850–2000 °С. Трещиностойкость образцов практически не меняется в ходе термобарического спекания. Формирование структуры образцов происходит без укрупнения зерен композита, что свидетельствует об отсутствии собирательной рекристаллизации.

***Ключевые слова**: аппарат высокого давления, сверхтвердая керамика, КНБ, фазовый состав, структура, титан*

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МАТЕРІАЛИ НА ОСНОВІ КНБ ЗІ ЗВЯЗКОЮ TiN-Al: СПІКАННЯ. СТРУКТУРА, ВЛАСТИВОСТІ

Методом спікання при високому тиску і високій температурі виготовлено матеріал на основі кубічного нітриду бору з використанням нітриду титану та алюмінію як зв'язувальної фази. Об'єкт дослідження — структура, фазовий склад та фізико-механічні властивості матеріалу. Встановлено, що при температурах спікання вище 1750 °С в результаті взаємодії між компонентами шихти для спікання відбувається утворення дибориду титану TiB_2 та AlN. Найвищі значення мікротвердості демонструють зразки, виготовлені в температурному інтервалі 1850 – 2000 °С. Тріщиностійкість зразків практично не змінюється в ході термобаричного спікання. Формування структури зразків відбувається без укрупнення зерен композиту, що свідчить про відсутність збиральної рекристалізації.

***Ключові слова:** апарат високого тиску, надтверда кераміка, КНБ, фазовий склад, структура, титан*

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INFLUENCE OF REINFORCEMENT BY THE WHISKERS OF Si_3N_4 AND $Mg_2B_2O_5$ ON THE PROPERTIES OF cBN-BASED COMPOSITES

Three types of cBN-based composites (without whiskers, reinforced with whiskers of Si_3N_4 and reinforced with whiskers of $Mg_2B_2O_5$) have been obtained by High Pressure-High Temperature (HPHT) sintering. Density, Young modulus, hardness, Poisson ratio and fracture toughness have been measured for all samples. cBN-based composites, that were reinforced by the whiskers of Si_3N_4, are characterized by better mechanical properties (hardness, fracture toughness) than non-reinforced cBN-based composites. But reinforcement by $Mg_2B_2O_5$ whiskers was insufficient, because $Mg_2B_2O_5$ whiskers have low thermochemical stability.

***Keywords**: whiskers, reinforcement, cBN, fracture toughness*

Introduction

Despite the development of high-performance conventional technology of forming (eg, injection molding, laser machining), cutting materials stay the most universal and popular way of forming machine-building products. It is well-known that during operations the cutting tool is subjected to the influence of temperature and pressure, which causes wear. Therefore, in the perfect case, the material of its cutting part of the tool must meet many requirements: have high hardness, durability, wear resistance, heat resistance, crack resistance (fracture toughness), adhesion resistance and cyclic strength, thermodynamic strength, heat capacity, thermal conductivity, low affinity for machining material, etc. cBN-based materials occupied a special place among the cutting materials due to such characteristics, like high strength, chemical stability over a wide temperature range [1]. HPHT sintering is traditional preparation method of cBN-based materials, which allow to obtain high-density materials with fine microstructure (consequently, these materials have high level of hardness and fracture toughness). Despite on this, the fracture toughness BL group stays unsatisfactory (about 2,5–5 $MPa \cdot m^{1/2}$, which is significantly lower than the fracture toughness of BH group – 9–10 $MPa \cdot m^{1/2}$) and this leads to a reduction in the service life of tools based on them [2–3]. Therefore, the problem of increasing the fracture toughness of this group of materials becomes obvious while maintaining the chemical stability of these materials [4]. One of the well-known ways to solve this problem is whisker reinforcement [5], which was widely used for other groups of cutting materials, but almost never used for this group [6].