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Preparation of materials based on Ti-Si-C system using high temperature – high pressure method

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Abstract: A method for densification of Ti_3SiC_2 powder (the *SHS* product) in High Temperature – High Pressure (*HT-HP*) Bridgman type apparatus was used. Compacts were sintered at pressure of 4.0±0.2 GPa and temperature about 1400°C for one, two and three minutes. The results of X-ray phase composition analysis of *SHS* powders were confronted with the phase analysis of obtained compacts. Some of physical and mechanical properties of Ti_3SiC_2 compacts were presented and compared to the properties of compacts prepared *HIP* method. *HT-HP* sintered compacts had very high hardness but low fracture toughness. This material did not show plastic behavior similar to hot pressing 85%vol Ti_3SiC_2 with 15%vol. The *HP–HT* technique always introduces strong structural stresses in compacts, which might result in cracks and even in their self-fragmentation. High hardness and sinterability pretend this material for the binder application in polycrystalline diamond compacts (*PCD*).

Keywords: *SHS* ceramic powders; Ceramic densification; *HT-HP* sintering; Properties

1. INTRODUCTION

Ti_3SiC_2 was first synthesized by Jeitschko and Novotny by chemical reaction between TiH_2, Si and graphite at 2000°C. The crystal structure is hexagonal corresponding to the D^4_{6h} ($P6_3/mmc$) space group and is composed of planar layers linked together by TiC octahedral [Ti_6C], they are separated by silicon layers. Its theoretical density is 4.53 g/cm^3. Its melting point to be in excess 3000°C.

Ti_3SiC_2 was synthesized for example by chemical vapor deposition techniques and for it preparation used self-propagating high-temperature synthesis [1, 2].

Polycrystalline bulk samples of Ti_3SiC_2 were fabricated by reactively hot – pressing Ti, graphite and SiC powders at 40 MPa and 1000°C for 4h. Its compressive strength, measured at room temperature was 600MPa. The room temperature electrical conductivity was 4.5x $10^{-6}K^{-1}$, with hardness of 4-6 GPa and a Young's modulus of 320 GPa [3]. Its combines typical ceramic properties like high stiffness, thermal and chemical resistance with very low hardness 4-6 GPa, high fracture toughness and ductile behavior under stress [3].

Self – propagating high temperature synthesis can be used for preparation of wide range of sinterable refractory ceramic powders. The powders (SiC, Si_3N_4, sialons, etc) can be pressureless sintered or hot-pressed into dense polycrystals [4]. A certain drawback of *SHS* technique is that the combustion products except the main phase usually contain also other minor phases.

J.Lis *et al.* obtained materials based on Ti_3SiC_2 by hot isostatic pressing of Ti_3SiC powders prepared by *SHS* method. They sintered dense polycrystalline materials with controlled content of hexagonal Ti_3SiC_2 phase. Reactions in ternary Ti-Si-C system under the *SHS* regime may produce either TiC, SiC, $TiSi_2$, Ti_5Si_3C or Ti_3SiC_2; each in quantity depending on the starting mixture composition. They realized the pressureless sintering and hot pressing of Ti_3SiC_2 (with presence of TiC phase). The hot – pressing allows to reduce sintering temperature and time and in case the grains growth was limited. Material was isostaticaly pressed under 350 MPa and sintered in a graphite resistance furnace in the argon atmosphere at 1470°C for 4 hours. This material had hardness *HV* up to 9.5±0.5 GPa, high fracture toughness 10.0±1.2 $MPam^{-1/2}$ and behavior at room temperature that was close to plastic materials [5]. A titanium silicon carbide Ti_3SiC_2 was reported to have interesting properties in between ceramics and metals, explained by its laminar structure [3,5]. Specifically, Ti_3SiC_2 combines high stiffness characteristic for ceramic materials with some ductile behavior characteristic for metals. Additionally, its specific heat, thermal and electric conductivity is closer to titanium metal than to titanium carbide [4, 5]. This hexagonal Ti_3SiC_2 is most probably a low-temperature phase stable below 1400-1450°C [6]. Nickl *et al.* indicated that while Ti_3SiC_2 formed by powder metallurgical processes did not show plastic behavior, their vapor-deposited compound did show unusual plastic behavior similar to graphite. The high stiffness in connection with some ductility and high penetration properties of this material makes it a prime candidate for superior bonding material for *PCD* compacts [1].

The aim of this investigations was to densification of Ti_3SiC_2 powder in High Temperature – High Pressure Bridgman type apparatus and analyze possibility of application the *SHS* synthesized Ti_3SiC_2 powders as binding phase for *PCD*.

2. EXPERIMENTAL PROCEDURES

The Ti_3SiC_2 were produced at Department of Advanced Ceramic AGH from stoichiometric mixtures of powders Ti, Si, C powders using *SHS* method. The titanium metal powder (GoodFellow) had particles below 150μm and 99.9% chemical purity. The silicon powder was a semi-conductor product of chemical purity 99.5%, it had particle size d_{BET} 5.15μm. A Merck Co. graphite powder of particle size below 50μm was used.

The homogeneity of these mixtures were ascertain through extensive mixing of their components for 12 hours in a vibratory rotary mill with Teflon balls suspended in anhydrous isopropyl alcohol. Drying was carried out during mixing. Next, powders were formed into discs by pressing in steel matrix and synthesized using *SHS* technique. The reaction was started at 1250°C using graphite crucible with the graphite foils lining in an argon-filled chamber. After ignition, i.e. rising part of material to high temperature, the front of reaction propagated across the reaction crucible resulting in full transformation of the loaded material. The products of *SHS* reaction were crushed in Abbich mortar to powder with grain size to 0.5mm and next they were milled in rotary-vibratory mill with WC grinding media in anhydrous isopropyl alcohol to power with specific surface area to $10m^2/g$.

The resulting *SHS* product of mainly Ti_3SiC_2 was formed into discs (Φ=15mm, h=5mm) by pressing in a steel matrix under pressure of 200 MPa. Samples were heated using an

assembly provided with an internal graphite heater. Compacts were sintered at pressure of 4.0±0.2 GPa and temperature about 1400°C in the high pressure Bridgman type apparatus. The durations of HT-HP sintering were 1, 2, 3 minutes.

The phase composition of *SHS* powders and Ti_3SiC_2 compacts were studied using X-ray diffractometer. Samples for Vickers hardness measurements and microstructure analysis were prepared through lapping on a cast iron plate with diamond paste. Density was measured using Arhimedes method. Hardness was measured with Vickers apparatus at 98 N load. Fracture toughness was measured by indentation by Vickers method with Palmquist cracks. Young's modulus were measured with the ultrasonic method using the ZVUK – 107 apparatus.

3. RESULTS

The X-ray analysis of Ti_3SiC_2 powder (Fig. 1) show that this *SHS* product is a complex mixture of $TiSi_2$, TiC and the main phase is about 85% vol. of Ti_3SiC_2. In Figure 2 is shown comparison of X-ray patterns of Ti_3SiC_2 powder and Ti_3SiC_2 compact from *HT-HP* sintering. Phases composition of powder and compact are very similar. There is only difference of picks intensity. There is not thermal decomposition of Ti_3SiC_2 during *HT-HP* sintering process.

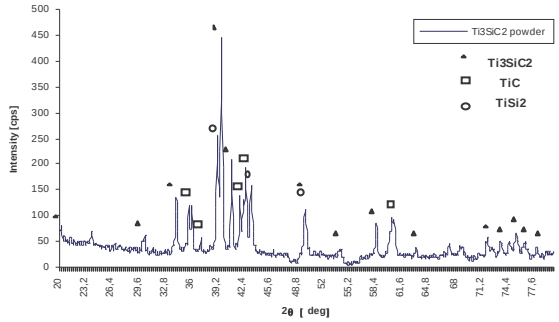

Figure 1. X – ray diffraction pattern of Ti_3SiC_2 powder

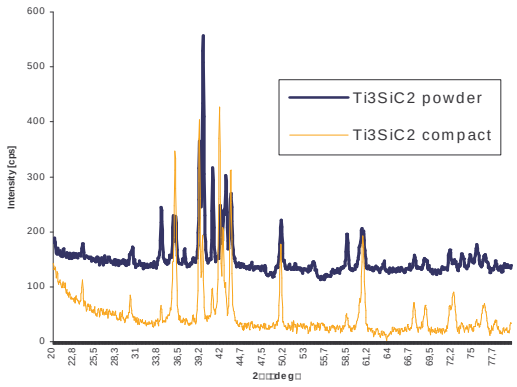

Figure 2. The comparison of X – ray diffraction patterns of Ti_3SiC_2 powder and Ti_3SiC_2 (*HT – HP* sintering)

The hardness of the Ti_3SiC_2 compacts strongly depend on duration of the sintering. The best result was obtained for 3 minutes of sintering. For compacts sintered for one and tree

minutes hardness *HV* are respectively 12 GPa and 14,1 GPa. Properties of the Ti_3SiC_2 compacts are summariesed in Table 1.

Table 1.
Selected properties of Ti_3SiC_2 obtained by *HP-HT* and *HIP* methods.

Properties	Ti_3SiC_2 compact obtained by *HP-HT* method	Hot pressing of mixtures: Ti_3SiC_2 85%vol., TiC 15%vol. [5]
ρ, g/cm^3	4.3	4.57
K_{Ic}, MPa m$^{-1/2}$ (HV10)	2.81 ± 0.22	10 ± 1.2
E, GPa	216	364 ± 0.4
HV10, GPa	12 ± 1.1÷14.0 ± 0.3	10 ±1.2

4. CONCLUSIONS

SHS in powders mixed with composition of 3Ti+Si+2C resulted in a complex material with Ti_3SiC_2, TiC and $TiSi_2$ phases. During very short duration of the *HT-HP* sintering the phase composition almost did not change. This material has very high hardness but low fracture toughness. This material did not show plastic behavior similar to hot pressing of 85%vol. Ti_3SiC_2 with 15%vol. TiC. The same tendency occurred for SiC compacts, sintered *HT–HP* method [7]. Probably this low fracture toughness is connected with stresses, which exist during *HT-HP* sintering. The *HT–HP* technique always introduces strong structural stresses in compacts, which might result in cracks and even in their self-fragmentation. High hardness and sinterability pretend this material for the binder application in polycrystalline diamond compacts (*PCD*).

REFERENCES

1. J.Nickl, K.K. Schweitzer, P. Luxenberg, Gasphasenabscheidung im systeme Ti-C-Si, J.Less-Common Met., no 26, pp. 283, 1972.
2. R.Pampuch, J.Lis, L.Stobierski, M.Tymkiewicz, Ti_3SiC_2-based materials produced by self propagating high – temperature synthesis (*SHS*) and ceramic processing, J.Mater. Synth.Process., no 1, pp. 93, 1993.
3. M.W. Barsoum, T. El-Raghy, Synthesis and characterization of remarkable ceramic: Ti_3SiC_2, J. Am. Soc., Vol. 79, no7, pp. 1953-1956, 1996.
4. J.Lis, Y. Miyamoto, R.Pampuch, K. Tanihata, Ti_3SiC_2 materials prepared by *HIP – SHS* techniques, Materials Letters, no 22, pp. 163-168, 1995.
5. T.Rudnik, J.Lis., The Ti_2SiC_2 based structural ceramics, Archives of Metallurgy, vol. 42, no1, pp. 59-66, 1997.
6. R.Pampuch, J.Lis, Ti_3SiC_2 – a plastic ceramic material, Ceramics: Charting the Future: edited by P.Vincenzini (Techna Srl, Faenza), pp. 725-732, 1995.
7. T. Gibas, L. Jaworska, Wysokociśnieniowe spiekanie węglika krzemu – możliwość zastosowania do celów narzędziowych, II Ogólnopolska konferencja naukowa „Problemy jakości symulatorem rozwoju technologii bezodpadowych, Metalurgia Proszków", Kraków 16 – 18. IX 1999, tom II, pp. 85-92, Politechnika Krakowska 1999.