SYNTHEISIS OF THE ZrB_2-SiC ULTRA-HIGH TEMPERATURE CERAMIC POWDER BY PLASMA SPHEROIDIZATION

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Abstract

The mixture of ZrB_2 and SiC powders were used as a raw material. The powders were milled in a planetary ball mill, blended with polyvinyl alcohol, and sieved to 0-125 µm. After sieving, the powder was spheroidized in Tekna Tek 15 ICP plasma plant. During the spheroidization, the ceramic powder melts and form a compound, containing ZrB_2-SiC and rapidly crystallized forming dense spherical shaped powders, suitable for additive manufacturing or sintering techniques.

Keywords: UHTC, additive manufacturing, powders, plasma spheroidization

1. INTRODUCTION

Ultra-high temperature ceramics (UHTCs) are candidate materials for a variety of aerospace applications owing to their unique combination of properties, including high melting temperature (>3273 K), high strength, and high elastic modulus [1–6]. Among them, Zirconium diboride (ZrB_2) is an outstanding material owing to its high melting temperature (>3000K), low theoretical density (6.09 g/cm3), high thermal conductivity (65-135 W/mK) and relatively low thermal expansion coefficient [7]. Above 1400°C, however, the oxidation resistance of pure ZrB_2 is very poor due to the volatilization of B_2O_3 and a residual porous ZrO_2 layer [8]. Adding SiC can significantly improve the oxidation resistance of ZrB_2-based coatings in the moderately high temperature range by the formation of borosilicate glass [9].

As a result of its strong covalent bond and low self-diffusion coefficient, it is challenging to fabricate the nearly fully dense ZrB_2-based ceramic or dense ZrB_2-based coating [10,11]. For sintering the ZrB_2-SiC ceramic, the main preparation technologies include hot pressing, reactive hot pressing, pressureless sintering, and spark plasma sintering (SPS) [12–15]. Until now, to further improve the densification of the ZrB_2-SiC ceramic, adding sintering aids and adjusting sintering parameters have been tried in several previous works. As for the ZrB_2-SiC coating, many methods, including plasma spraying [16], vapour silicon infiltration (VSI) [17], painting slurry [18], in situ reaction [19], and pack cementation [19]. Wang et al.[20] designed a gradient structure and prepared a dense and defect-free SiC-ZrB_2-$MoSi_2$ coating on SiC coated C/C composites using supersonic plasma spraying. Lietal. [21] introduced a relatively low melting point WB into ZrB_2-SiC coatings to improve the densification and oxidation resistance of the coating. The eutectic ZrB_2-SiC composition powder were obtained by plasma spheroidization for spray spraying [22].

In this work the results of synthesis of the ZrB_2-SiC spherical powders from elemental powder mixture by plasma spheroidization are presented.

2. MATERIALS AND METHODS

The elemental powders of ZrB_2 and SiC were used as raw materials. The powder mixture of 80ZrB_2-20SiC (vol%) was blended in planetary ball mill Fritsch Pulverisette 4 with the addition of 5 (vol%) of the Polyvinylalcohol (PVA) as a binder reagent. After the milling, the mixture was sieved into the 0-125 μm fraction with subsequent plasma spheroidization in 15 kW ICP plasma torch (by Tekna Plasma Systems inc.) with powder feed rate 4 g/min in argon-hydrogen plasma. The phases were analyzed by XRD Bruker D8 ADVANCE. The microstructure of samples was characterized using SEM Mira3 Tescan. The chemical elements distribution was studied by X-ray elemental mapping.

3. RESULTS AND DISCUSSION

Figure 1 shows the ZrB_2-SiC phase diagram [6]. The addition of the SiC leads to a decrease in the melting temperature and form the eutectic composition. The XRD results (**Figure 2**) shows the diffraction peaks of the SiC, ZrB_2 and the presence of the Si and C. The latter may be the signs of the dissociation of the SiC under the high-temperature plasma treatment.

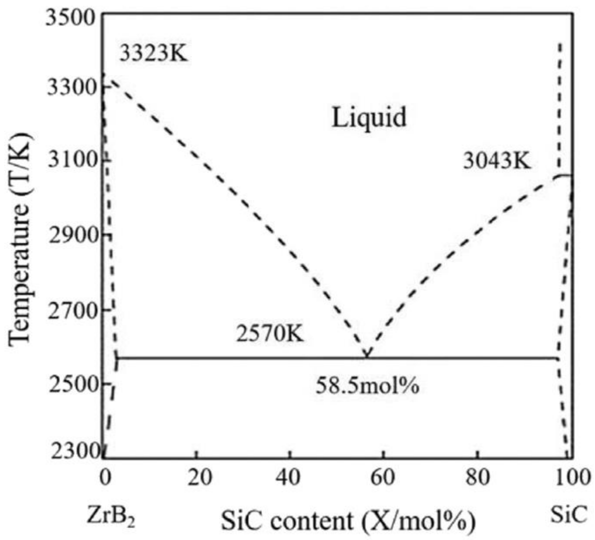

Figure 1 SiC-ZrB_2 phase diagram [6]

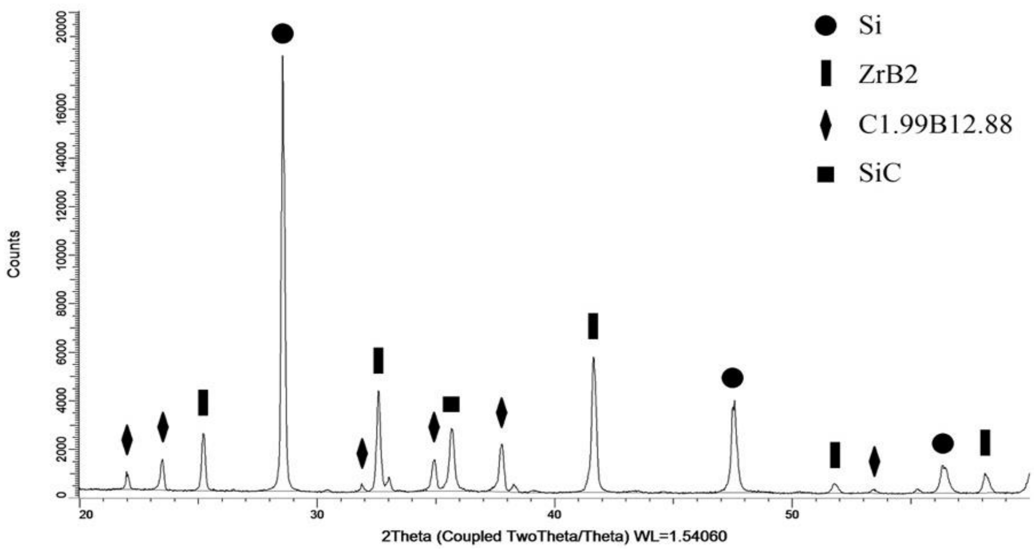

Figure 2 XRD pattern of the powder after plasma spheroidization

The morphology of the powders after plasma spheroidization is presented by near-ideal spherical shape particles with the presence of nanoparticles that agglomerated around the particles due to the electrostatic interaction (**Figure 3**). Those nanoparticles are the result of overheating, vaporizing and condensing of the smaller particles in the fraction and the decomposition of the PVA with the resulting carbon nanoparticles. On the surfaces of the particles, it can be noticed the signs of the epitaxial growth of the ceramics.

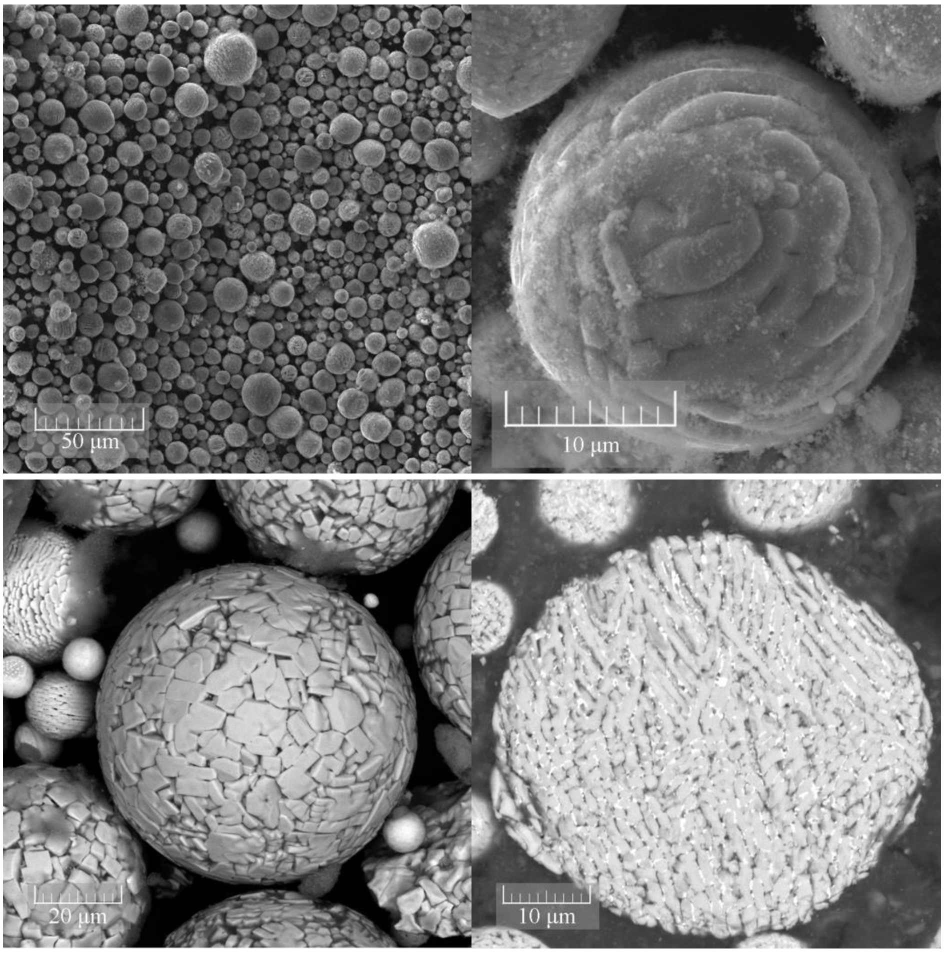

Figure 3 SEM micrographs of the powder after plasma spheroidization

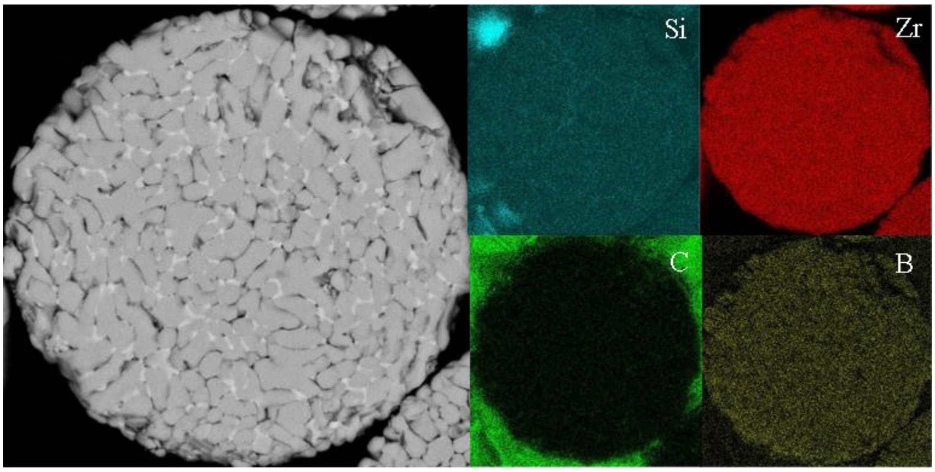

Figure 4 Elemental mapping of the powder particle after plasma spheroidization

The elemental mapping (**Figure 4**) shows the distribution of the chemical elements in the powder particle volume. The distribution is relatively uniform across the powder particle. The microstructure consists of the lamellar eutectic-type structure with contrasting phases of SiC+ZrB_2. It can be seen, the majority of the particles are dense and there are no signs of the hollow particles, which means in addition to the spherical morphology and fraction, that the plasma spheroidized ZrB_2-SiC powder meets the requirements for the additive manufacturing technologies, such as binder jetting.

4. CONCLUSION

In the present work, the dense spherical powders of the mixture 80ZrB_2-20SiC (vol%) were obtained by mechanical mixing and plasma spheroidization.

The microstructure of the powder consist of the uniformly distributed lamellar-eutectic type of structure with contrasting phases, which in accordance with XRD can be determined as ZrB_2+SiC. There also a noticeable amount of nanoparticles as a result of the decomposition of the organic binder and smaller particles due to the overheating in plasma.

The spherical shaped dense particles can further be used for additive manufacturing technologies such as binder jetting.

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