Fusion Engineering and Design 103 (2016) 118–124

Contents lists available at ScienceDirect

Fusion Engineering and Design

journal homepage: www.elsevier.com/locate/fusengdes

Composition and microstructure of beryllium carbide films prepared by thermal MOCVD

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HIGHLIGHTS

- Non-columnar-crystal Be_2C films were firstly prepared by thermal MOCVD.
- Beryllium carbide was always the dominant phase in the films.
- α-Be and carbon existed in films deposited below and beyond 400 °C, respectively.
- Morphology evolved with temperatures and no columnar grains were characterized.
- The preferred substrate temperature for depositing high quality Be_2C films was 400 °C.

ARTICLE INFO

Article history:
Received 26 September 2015
Received in revised form
25 November 2015
Accepted 8 December 2015
Available online 6 January 2016

Keywords:
Beryllium carbide
Capsule materials
Diethylberyllium
Thermal MOCVD
Inertial confinement fusion

ABSTRACT

Beryllium carbide films without columnar-crystal microstructures were prepared on the Si (1 0 0) substrate by thermal metal organic chemical vapor deposition using diethylberyllium as precursor. The influence of the substrate temperature on composition and microstructure of beryllium carbide films was systematically studied. Crystalline beryllium carbide is always the dominant phase according to XRD analysis. Meanwhile, a small amount of α-Be phase exists in films when the substrate temperature is below 400 °C, and hydrocarbon or amorphous carbon exists when the temperature is beyond 400 °C. Surfaces morphology shows transition from domes to cylinders, to humps, and to tetraquetrous crystalline needles with the increase of substrate temperature. No columnar grains are characterized throughout the thickness as revealed from the cross-section views. The average densities of these films are determined to be 2.04–2.17 g/cm^3. The findings indicate the substrate temperature has great influences on the composition and microstructure of the Be_2C films grown by thermal MOCVD.

1. Introduction

There are concerns that energy shortage may increase in the future. It is believed that controlled nuclear fusion will provide an alternate and cleaner energy source for the future [1–3]. Inertial confinement fusion (ICF) is one of the most potential methods to achieve controlled fusion reactions [4]. In an ICF reactor, the fusion fuel (deuterium–tritium mixture) is confined inside a small capsule. The most principal requirements for capsule materials are low atomic number, uniform composition and structures, and high density [5–7]. Beryllium carbide is a potential capsule material [8] for its low average atomic number (Z = 4.7), relative high density (ρ = 2.4 g/cm^3), and oxidation resistance [9,10].

Beryllium carbide was first synthesized by Lebeau with two methods: (a) heating a mixture of beryllium and sugar charcoal, and (b) heating beryllium oxide with sugar charcoal. Then many researchers fabricated this carbide with Lebeau's methods and studied its properties [11–13]. Thereafter, Xie and Shih, etc. developed two methods (Plasma Polymerization of Diethyl-beryllium and Be sputtering into an Ar/Methane Plasma) to deposit beryllium carbide coatings for application to ICF [8,10,14,15]. The H_2 permeability, mechanical properties, chemical composition, and electrical and thermal conductivities of beryllium carbide films were analyzed. The results indicated most of the requirements of the ignition target were satisfied. However, those films also exhibited columnar crystal microstructures (similar to beryllium) [15,16], which will decrease the isotropic ablative characteristic and promote the

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http://dx.doi.org/10.1016/j.fusengdes.2015.12.015
0920-3796/

increasing of Rayleigh–Taylor instability, resulting in the failure of ignition [17–21]. The objective of the present work is to fabricate Be_2C films without columnar-crystal microstructure by thermal MOCVD, and to investigate the intrinsic relationship between the substrate temperature and the phase component, chemical composition, microstructure, and density of the films.

2. Experiment

Beryllium carbide films were deposited in a homemade, cold-wall vertical MOCVD reactor as shown in Fig. 1. Diethylberyllium $\{Be(C_2H_5)_2$, purity 99.9%$\}$ was used as the precursor, whose saturated vapor pressure is 1.6 Torr at room temperature. Si (100) chips (25 mm × 25 mm) were used as substrates. The distance between the substrate and outlet was set to be 50 mm. More process parameters for the deposition are summarized in Table 1. Before the deposition, the substrates were in situ annealed 30 min in hydrogen ambient at 500 °C in order to remove any residual surface contaminants. The influence of substrate temperatures on the composition and microstructure will be discussed.

The crystallinity and phase composition of the films were characterized by Grazing Incidence ($\theta = 0.5°$) X-ray Diffraction (GIXRD) [22] with Cu Kα (0.154056 nm) radiation, owing to its low atomic number resulted difficulty in detection by typical XRD. The use of glancing geometry improved the effective diffraction volume of the films and decreased the influence of substrates. To eliminate the influence of substrates, the depositions were rotated in the sample plane to a position where peaks corresponding to Si {1 0 0} were avoided. The film compositions were analyzed by Energy Dispersive X-ray Spectra (EDX) and X-ray (Mg Kα 1253.6 eV) Photoelectron Spectroscopy (XPS), as well as Infrared Carbon Element Analyzer (ICEA), and Inductively Coupled Plasma Mass Spectrometry (ICP-MS). Surface and cross-section morphologies were investigated with Scanning Electron Microscope (SEM). The average densities were calculated from the weight gain and the thickness (evaluated from SEM) of films with definite area.

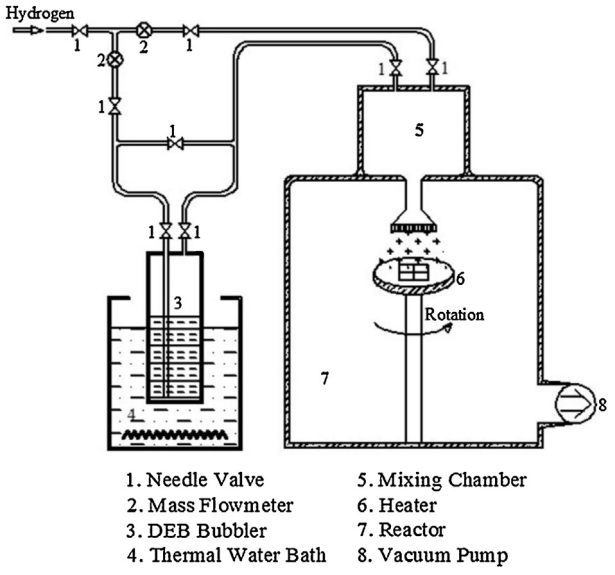

Fig. 1. Schematic diagram of the deposition system. (JPG format, 600 dpi, single-column fitting image)

Table 1
Process parameters for beryllium carbide films deposition.

Precursor	Diethyl-beryllium (DEB)
Working vacuum	~0.4 Pa
Background vacuum	~8×10^{-6} Pa
Vaporizer temperature (T_{vap})	25 °C
Carrier hydrogen flow rate (R_c)	5sc cm
Diluent hydrogen flow rate (R_d)	10sc cm
Substrate temperature (T_{sub})	300/350/400/470 °C
Deposition time	8 h

3. Result and discussion

3.1. Composition analysis

Fig. 2 shows the X-ray diffraction patterns of films with substrate temperatures of 300, 400, and 470 °C, by comparison with the standard Be_2C powder XRD spectra (PDF-33-0191) [23]. All strong peaks of the Be_2C, (1 1 1)/(2 0 0)/(2 2 0)/(3 1 1)/(3 3 1), can be found distinctly in the XRD patterns as labeled in Fig. 2, indicating that beryllium carbide is the dominant phase in these films. However, the diffraction peaks corresponding to α-beryllium are observed apart from the Be_2C peaks when substrate temperature is 300 °C, revealing that the film consists of crystalline beryllium carbide and α-beryllium. With an increase of the substrate temperature up to 400 °C, α-beryllium phase disappears, and Be_2C is the unique phase. The peak intensity ratio of $I_{Be2C(220)}/I_{Be2C(111)}$ for films deposited at different substrate temperatures are listed in Table 2. In randomly oriented Be_2C powders, the ratio of $I_{Be2C(220)}/I_{Be2C(111)}$ is equal to 0.75. Table 2 shows the preferred orientation of beryllium carbide changed from (1 1 1) to (2 2 0) with the increasing of T_{sub}. In particular, the film deposited at 400 °C exhibits an untextured structure, where the $I_{Be2C(220)}/I_{Be2C(111)}$ is 0.6. This could be associated with

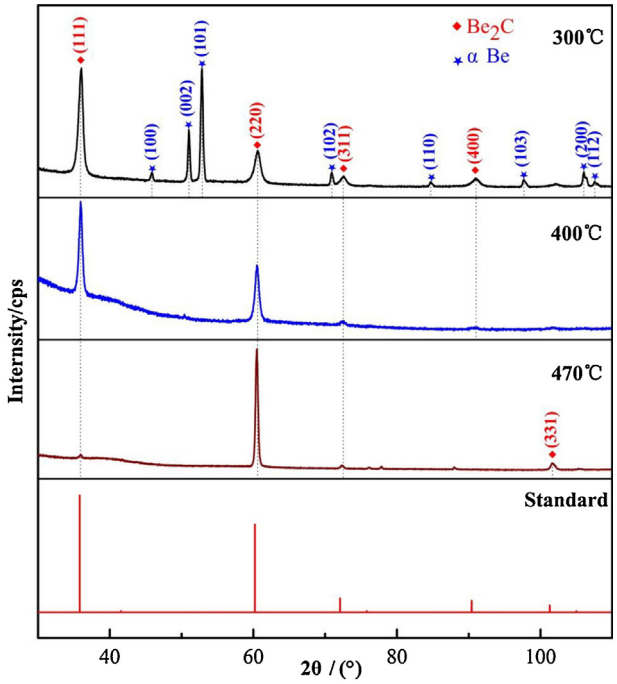

Fig. 2. XRD patterns of Be_2C coatings at different deposition temperatures. (JPG format, 1200 dpi, single-column fitting image)

Table 2
Thickness, Peak intensity ratio (I_{220}/I_{111}) and grain sizes of Be_2C coatings.

T_{sub} (°C)	Thickness (nm)	I_{220}/I_{111}	Grain size (nm)
300	2497	0.3	17.4
350	2648	–	–
400	6288	0.6	21.0
470	5952	34.7	25.6