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Preparation of TaC Coatings via Wet Powder Process

Shen Xiaosong, Wang Song, Li Wei, Zhang Jian

Science and Technology on Advanced Ceramic Fibres and Composites Laboratory, National University of Defense Technology, Changsha 410073, China

Abstract: The TaC coatings were prepared by precursor slurry application and subsequent sintering processes on graphite substrate. The phase composition and morphology of the prepared coating was investigated by XRD and SEM. The results show that the coating consists of single-phase TaC and has a dense granular structure containing grains with the mean size of 12 μm. The coating has an isotropic granular structure, which makes the crack tip be pinned at a grain boundary junction and the crack would not fully penetrate the coating. The hardness and the elastic modulus of the coating are 15.35 GPa and 195.1 GPa, respectively.

Key words: TaC precursor powders; sol-gel; TaC coating

TaC is a promising ceramic material, because it possesses attractive properties of high hardness, high melting point (3880 ℃), good resistance to chemical attack and thermal shock, good thermal and excellent electrical conductivity, and it shows good chemical compatibility with the graphite [1-5]. Compared with surface-carbonized bulk Ta components and bulk TaC ceramics, the TaC-coated components, such as TaC-coated graphite, are particularly versatile, especially in terms of manufacturing complex-shaped components, higher reliability and lower cost [6,7].

TaC coating resists chemical attack by corrosive liquids and gases at very high temperatures. There are increasing acceptance in sealing and protecting graphite hardware from hot ammonia, hydrogen, hydrochloric acid, and molten metal used in compound semiconductors epitaxial processes such as metal-organic chemical vapor deposition (MOCVD) and physical vapor transport (PVT)[8-16].

Herein, we proposed a method of coating TaC on graphite substrates. This method simultaneously provides low cost, high reliability and durability, a high degree of freedom in shape forming, and near-net-shape production. TaC coatings were formed via TaC precursor slurry (containing TaC precursor powders) application on graphite substrates and subsequent sintering processes.

1 Experiment

The TaC precursor slurry was a mixture of TaC precursor powder (synthesized by our laboratory), organic solvents, and binder. The TaC precursor slurry was applied to the graphite substrate with a spray gun or paintbrush, and preliminarily heated to a temperature of 423 K for 30 min to completely evaporate the organic solvents from the powder compact film. The TaC precursor powder compact film on the substrate was sintered at 2273 K in a vacuum atmosphere.

The phases present in the coatings were determined by X-ray diffraction with a Bruker D8. Cu Kα radiation (λ= 0.154 06 nm) was operated at 40 mA and 40 kV. A field emission gun scanning electron microscope (HITACHI FEG S4800, 10 kV) equipped with energy dispersive spectroscopy (EDS, 15 kV) was used to examine microstructure and elemental composition of the powders and coatings. The nano-indentation hardness and the elastic modulus of the coating were examined by the MTS Nano-indenter XP. The Atomic Force Microscope (Nanoman VS) were used to observe the morphology of indentation.

2 Results and Discussion

2.1 Microstructure of the TaC precursor powder

SEM analyses were used to obtain the information about the size and morphologies of the TaC precursor powders in Fig.1. The powders exhibit a sphere-like morphology and disperse uniformly. These particles present homogeneous grain size of about 1 μm, as shown in Fig.1a. The chemical composition judged by the EDS result is $Ta_{3.9}O_{7.3}C$ (Fig.1b), tantalum oxide (Ta_2O_5) in the TaC precursor powders would react with carbon (C) in the graphite substrate, which may improve the bonding strength between the coating and substrate.

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Corresponding author: Wang Song, Ph. D., Professor, Science and Technology on Advanced Ceramic Fibres and Composites Laboratory, National University of Defense Technology, Changsha 410073, P. R. China, Tel: 0086-731-84576441, E-mail: wangs_0731@163.com

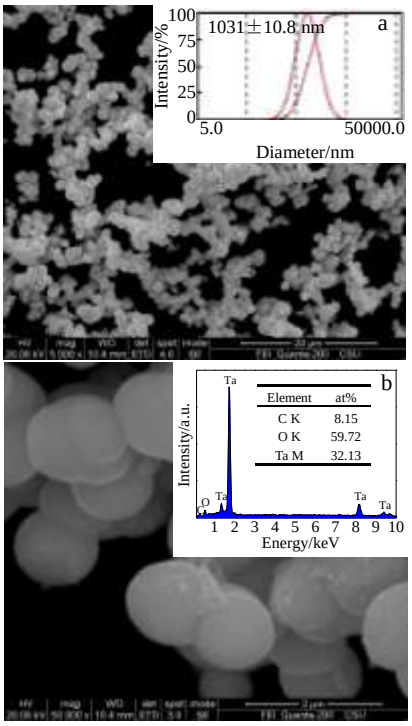

Fig.1 SEM images of the resulting TaC precursor powders: (a) powders morphology and grain size distribution; (b) EDS result of the powder ($Ta_{3.9}O_{7.3}C$)

2.2 Microstructure of the TaC coating

Fig.2 shows an example of the process for fabricating a TaC coating on graphite. Graphite material, shaped into a wafer, was used as the substrate for TaC coating, as shown in Fig.2a. Comparing Fig.2b and 2c, we can find that the TaC precursor powder compact film with a matte gray appearance becomes a dense film with a yellow-gold surface and a metallic-luster after sintering.

Fig.2 Processing sequence for TaC coatings on graphite wafer: (a) as-machined graphite wafer used as a substrate, (b) graphite wafer covered with a TaC precursor powder compact film by spray coating a TaC precursor slurry, and (c) resultant TaC coating on graphite formed by sintering the spray-coated wafer

The X-ray diffraction (XRD) analysis was carried out to obtain crystalline structure of the TaC coatings. The diffraction peak positions from the TaC coatings and the standard TaC are compared in Fig.3a. It shows that the coating consists of single-phase TaC with a rock-salt structure, and no other crystalline phases are detected. The peaks with 2θ values of 34.81°, 40.42°, 58.48°, 69.89°, 73.49°, and 87.39° correspond to the crystal planes of (111), (200), (220), (311), (222), and (400), respectively, for TaC phase. The lattice constants for the TaC coatings (Fig.3b) were determined by an extrapolation method based on the Nelson-Riley function. The lattice constant values obtained here is almost identical to those reported by Valvoda [17] (for TaC 0.99: 0.445 56±0.001 nm). It indicates the C/Ta ratio of the TaC coatings is close to 1.0, without a significant carbon deficit.

In addition to the crystal structure analysis by XRD, an SEM analysis was carried out to obtain morphologic information. The SEM image of the as-sintered TaC coating in Fig.4a reveals that the coating has a dense granular structure containing grains with the mean size of 12 μm. The chemical composition of the coating judged by the EDS result is $Ta_{0.95}C$, as shown in Fig.4a. The morphology of the fractured surface (Fig.4b) shows that TaC coating fractures predominantly

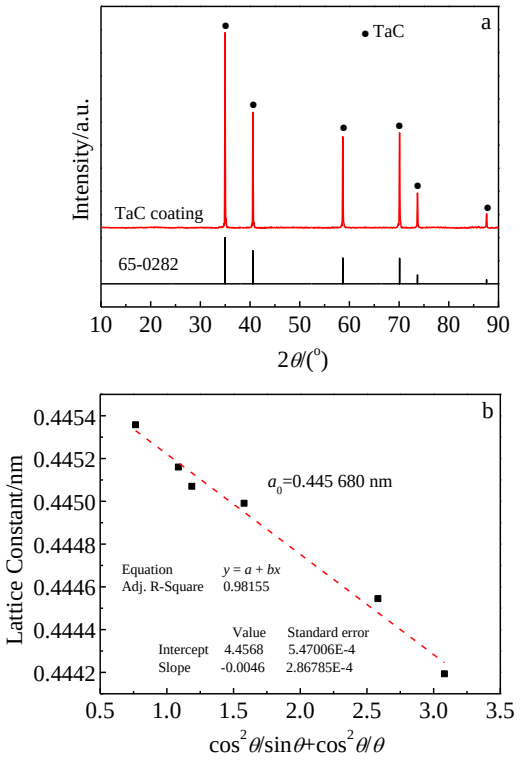

Fig.3 XRD pattern of the TaC coating (a); the lattice constants of TaC coating determined by an extrapolation method based on the Nelson-Riley function (b)

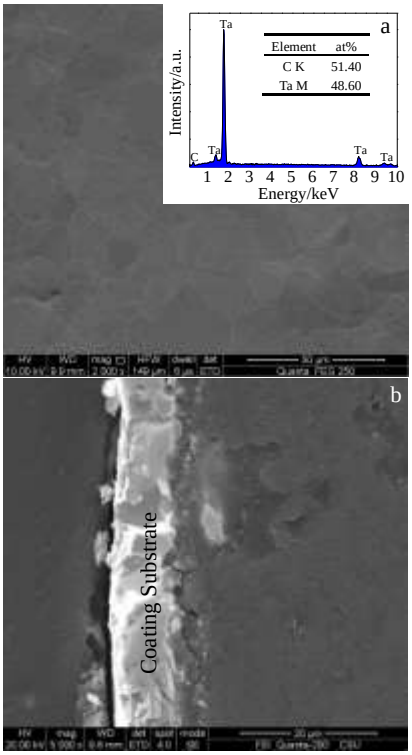

Fig.4 SEM images of TaC coating: (a) surface morphology and EDS result; (b) cross-sectional morphology

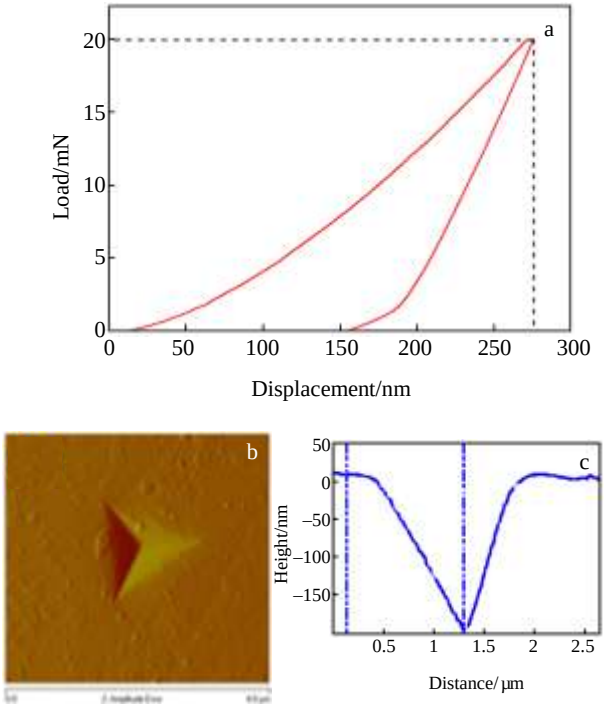

Fig.5 Load-displacement curves (a), AFM image of the indentation morphology (b) and the height-distance curve (c) of TaC coatings

at grain boundaries (smooth planes) and subordinately within grains (ridged planes). Furthermore, the coating has an isotropic granular structure. Fig.4b shows no cracks or pinholes penetrating through the coatings. Owing to this non-textured granular structure, even if there is some cracks due to thermal stress and/or heat shock in the TaC coating, the crack tip would be pinned at a grain boundary junction and would not fully penetrate the coating. These fatal defects in protective coatings are not present in TaC coatings which is an origin of the high reliability and durability of the coating.

2.3 Mechanical properties of the TaC coating

Nano-indentation was used to examine the hardness and the elastic modulus of the coating, Fig.5a is the load displacement curves and the image about the morphology of indentation. The values of nano-indentation hardness and the elastic modulus of the coating are 15.35 GPa and 195.1 GPa, respectively. As shown in Fig.5b and 5c, the surface of the coating is smooth and the indentation is clearly. There are neither pile-up nor sink-in surrounding of the indentation, which illuminates the result of the value are exact relatively.

Up to now, the study of hardness and elastic modulus of TaC have been focused on bulk TaC ceramics, and there were less research in the coatings or films. The results of hardness and elastic modulus of TaC ceramics from different researchers have biggish difference: the hardness of bulk TaC ceramics is in the range of 15~24 GPa[18,19], and the elastic modulus of bulk TaC ceramics is in the range of 241~722 GPa[20]. The origin of the lower elastic modulus of TaC coating prepared in this paper than reported in literatures may be attributed to the coarser grain size (about 12 μm) [21].

3 Conclusions

1) TaC coatings are prepared via apply TaC precursor slurry on graphite substrate and sintering subsequently.

2) The coating consists of single-phase TaC with a rock-salt structure, and it has a dense granular structure containing grain with the mean size of 12 μm. The coating has an isotropic granular structure, and shows no cracks or pinholes penetrating through the coatings. This non-textured granular structure makes the crack tip be pinned at a grain boundary junction and the crack would not fully penetrate the coating.

3) The values of nano-indentation hardness and the elastic modulus of the coating are 15.35 GPa and 195.1 GPa, respectively.

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湿粉末法制备 TaC 涂层

沈小松，王 松，李 伟，张 健
(国防科学技术大学 新型陶瓷纤维及其复合材料重点实验室，湖南 长沙 410073)

摘 要：通过在石墨基体涂覆前驱体料浆，在高温下烧结后得到 TaC 涂层。通过 XRD 和 SEM 对涂层形貌与结构进行表征，TaC 涂层由平均尺寸为 12 μm 的致密颗粒组成。涂层晶粒生长方式为自由取向生长，这种生长模式使得裂纹在扩展时会被钉扎在晶界连接处，而不会贯穿整个涂层。TaC 涂层的硬度和弹性模量分别为 15.35 GPa 和 195.1 GPa。

关键词：TaC 前驱体粉末；溶胶-凝胶；TaC 涂层

作者简介：沈小松，男，1992 年生，硕士生，国防科学技术大学新型陶瓷纤维及其复合材料重点实验室，湖南 长沙 410073，电话：0731-84576441，E-mail: m18073153758@163.com