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Microstructural formations and phase transformation pathways in hot isostatically pressed tantalum carbides

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Abstract

A series of XTa:$(1-X)$C ($0.5 < X < 1$) compositions have been fabricated by hot isostatic pressing (HIP) of Ta and TaC powder blends. Depending upon the targeted stoichiometry, single- or multiple-phase microstructures formed. The single-phase microstructures of both TaC and Ta_2C had equiaxed grain morphologies. The multiphase microstructures had either equiaxed TaC grains with a crisscross pattern of Ta_4C_3 laths or acicular grain morphologies with rafts of TaC, Ta_4C_3 and Ta_2C laths running parallel to the major axis of the grains. The effect of phase transformations on the microstructure of these specimens is discussed and compared to those microstructures seen in a reaction diffusion couple formed between Ta and TaC powders processed under the same HIP conditions. This couple revealed the depletion of carbon from the TaC phase and its reaction with the tantalum metal to form the various Ta-rich carbide phases. The precipitation sequence was found to be paramount in controlling the grain morphology. A close-packed plane and direction orientation relationship was seen between all the phases. The crisscross pattern of Ta_4C_3 precipitation in TaC was a consequence of TaC's multiple variant {1 1 1} orientations and had little or no effect on the grain morphology. In contrast, the single variant close-packed plane {0 0 0 1} in Ta_2C resulted in the parallel alignment of the precipitated phases within its grain and an anisotropic growth direction that facilitated the acicular grain morphology.

Keywords: Ceramics; Carbides; Transmission electron microscopy (TEM); Electron backscattering diffraction (EBSD); Precipitation

1. Introduction

Tantalum carbide (TaC) has high hardness and a melting temperature of ~4250 K, which is one of the highest melting points known [1]. Consequently, tantalum carbide has been proposed for use in ultrahigh-temperature load-bearing applications [2–4], such as thermal barrier coatings, machine tools and wear-resistant brake liners. The phase diagram for the tantalum–carbon system [5] is shown in Fig. 1a with corresponding crystal structures shown in Fig. 1b. The ultrahigh melting temperature of TaC and the ability to precipitate similar high-melting-temperature phases offers the potential to engineer its microstructures to meet specific thermal–mechanical property combinations [6].

The TaC phase is a B1 compound (NaCl-based or rock-salt structure), with carbon atoms occupying the octahedral interstitial sites in a tantalum face-centered cubic (fcc) lattice [2,7]. The carbon-deficient Ta_2C phase has a melting temperature of ~3600 K [8,9] and consists of hexagonal metal layers separated by either an α-ordered or β-disordered carbon sublattice. The allotropic phase transformation temperature between α-Ta_2C (CdI_2 antitype structure) and β ($L'3$ structure) is ~2300 K [1,7]. In addition, a ζ-Ta_4C_3 phase, which is rhombohedral with the space group R_3m, can form at ~2775 K [10,11]. The Ta_6C_5 ordered cubic phase (Fig. 1a) represents an ordering of the vacancies on the carbon sublattice, and is thought to form during slow continuous cooling after annealing at

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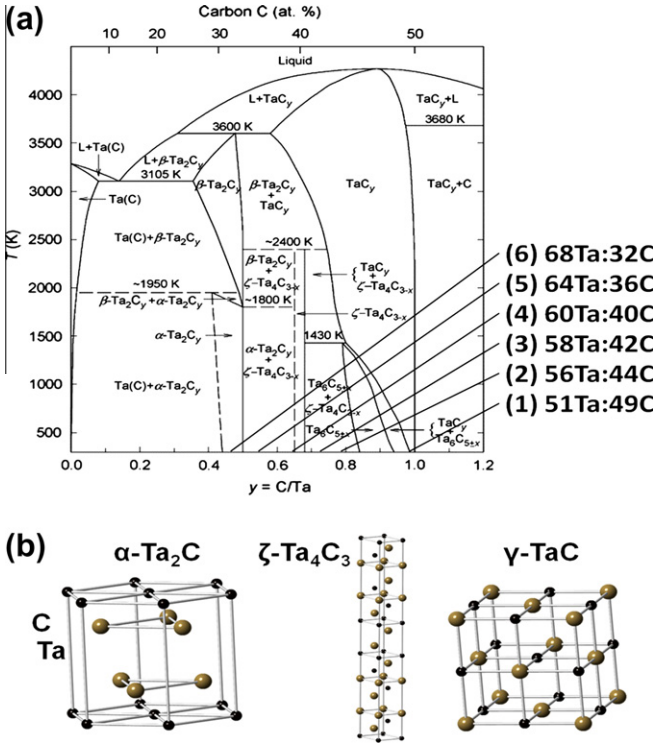

Fig. 1. (a) Ta–C binary phase diagram [9], with the region of the Ta/C content investigated highlighted. Numbers in parentheses identify compositions under investigation and correspond to the samples in Tables 1 and 2. For this work, the Ta_6C_5 regions are considered to be TaC. (b) The three phases of tantalum carbide: hexagonal α-Ta_2C, rhombohedral ζ-Ta_4C_3 and cubic γ-TaC.

elevated temperature [10]. Based on the thermal history of this work, the phase fields designated Ta_6C_5 are considered to be TaC.

One of the significant challenges of developing tantalum carbide alloys and other similar refractory compounds is their high melting points, which limits easy manufacturing to full density and near net shape. Additionally, the carbide systems can decarburize in a vacuum or inert gas atmospheres, which provides a challenge in controlling stoichiometry. This decomposition by incongruent vaporization is seen in the top-down solidification approaches, such as vacuum plasma spray and arc melting [12]. In hot isostatic pressing (HIP), the constituent powders themselves can still volatilize; however, this is minimized within the confines of the HIP can, thus providing process control of the Ta/C molar ratios. Thus a well-blended mixture of powders can reduce or eliminate significant compositional gradients in the final product. Finally, significant plastic flow occurs in the HIP billet, because of the high temperatures and pressures, and this yields near-theoretical densities [13]. In this paper, we report how HIP processing of tantalum carbides with varying compositions affects the precipitation of the different carbide phases and how this affects the resulting microstructure. Previous work has shown critical links between the strength and toughness in tantalum carbides to the volume fraction of phases formed with their corresponding microstructures [6,14]. By understanding the sequence of precipitation, the overall microstructures can ultimately be engineered. To date, there has been little work elucidating how these microstructures develop.

2. Experimental procedures

Two sets of specimens were prepared in order to study the precipitation and formation pathways of tantalum carbides. A set of six HIPed specimens of various Ta/C ratios, XTa:$(1-X)$C, where $X = 51$, 56, 58, 60, 64 and 68 at.%, were prepared by mixing powders of Ta and TaC to nominal compositions, as shown in the phase diagram of Fig. 1a and tabulated in Tables 1 and 2. In addition, a reaction diffusion couple specimen was formed by packing TaC powder onto Ta powder and HIPing under the same conditions as the blended specimens. The compositions produced in the couple included those of the individual specimens and was used to help explain the microstructures that formed from the single composition mixtures. The powders were purchased from Cerac, Inc., and their starting size distribution (Fig. 2) was measured from scanning electron microscopy images in conjunction with the Nikon Elements® imaging analysis software package. The grains sizes were measured using the diameters equivalent area circles, and typically encompassed 50–100 grains per specimen. The powders were placed into tantalum HIP cans using an inert-gas glove box and the tantalum cans were then evacuated and welded closed for subsequent HIPing at 200 MPa in an argon atmosphere at 1873 K for 100 min.

Table 1
The far left column contained the targeted atomic weight compositions. Grain size analysis data for the XTa:$(1-X)$C specimens.

Samples/grain size	Mean diameter (μm)	Mean long axis (μm)
(l) 51Ta:49C	2.2 ± 0.7	2.8 ± 0.9
(2) 56Ta:44C	5.2 ± 2.0	7.0 ± 4.0
(3) 53Ta:42C	8.9 ± 3.6	11.9 ± 4.0
(4) 60Ta:40C	9.6 ± 5.0	18.0 ± 11.1
(5) 64Ta:36C	6.1 ± 4.0	11.0 ± 7.7
(6) 68Ta:32C	23.0 ± 8.8	29.0 ± 10.8

Table 2
The far left column contains the targeted atomic weight compositions, the XRD is the estimated volume fraction of the phases determined by the integrated intensity ratio of the peaks from Fig. 3f, and the lever rule is the compositions based on the XRD volume fraction estimations.

Samples/phase	XRD			Lever rule	
	TaC	Ta_4C_3	Ta_2C	Ta	C
(1) 51Ta:49C	100	0	0	51	49
(2) 56Ta:44C	100	0	0	56	44
(3) 58Ta:42C	96	4	0	58	42
(4) 6QTa40C	49	33	17	61	39
(5) 64Ta36C	7	36	57	65	35
(6) 63Ta32C	0	0	100	68	32