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Ultrahigh-pressure consolidation and deformation of tantalum carbide at ambient and high temperatures

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Abstract

The deformation mechanism of the ultrahigh-temperature ceramic, tantalum carbide (TaC), consolidated at room temperature at a very high hydrostatic pressure of 7.7 GPa is investigated using high-resolution transmission electron microscopy. The deformation behavior of TaC at room temperature is also compared with that consolidated at high temperature (1830 °C) at a similar pressure. TaC could be consolidated to a bulk structure (90% theoretical density) at room temperature. The deformation mechanisms operating at room temperature and 1830 °C are found to be significantly different. The room-temperature deformation is dominated by the short-range movement of dislocations in multiple orientations, along with nanotwinning, grain rotation, crystallite misorientation with low-angle grain boundary formation and lattice structure destruction at interfaces. In contrast, at high temperature, the strain is accommodated mostly by a single slip system, forming a parallel array of dislocations. The consolidation at room temperature occurs by heavy deformation with the support from short range diffusion, whereas the consolidation at high temperature is mostly diffusion dominated, indicating a classic sintering mechanism. The improved degree of consolidation with fewer defects results in significantly improved elastic modulus and hardness in the case of high-temperature consolidate.

Keywords: Tantalum carbide; Ultrahigh pressure; Room-temperature consolidation; Deformation mechanism

1. Introduction

Tantalum carbide is an important transition metal monocarbide with an extremely high melting point of 3880 °C [1]. TaC has shown great promise as the next-generation aerospace material for throat and nozzle inserts due to its excellent refractory nature that can withstand the combustion flame temperature for most propellants [2]. However, the high melting point of TaC has made its consolidation into useful engineering shapes a major challenge. Achieving a fair degree of consolidation requires high-temperature exposure, as sintering occurs typically at temperatures $>0.6T_m$. The temperature required for consolidation is usually >2200 °C in pressureless conditions, whereas in pressure-assisted sintering (spark plasma sintering or hot isostatic pressing) it is >1200 °C [3–7]. Consolidation of TaC has been performed using different techniques, e.g. pressureless sintering, hot pressing, hot isostatic processing, high frequency induction heating, vacuum plasma spraying, spark plasma sintering and dynamic consolidation [8–21]. Additives like boron carbide, hafnium carbide, niobium carbide etc., have effectively reduced the sintering temperature to as low as 1400 °C, but at the expense of structural fineness and loss of mechanical performance [5,7]. Thus, there is a need for a suitable consolidation

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technique for TaC which can effectively decrease the consolidation temperature and retain the fine grain structure simultaneously.

The application of high pressure aids in consolidation through the powder metallurgy technique. Gallas and co-workers' method of consolidating nanosized ceramic particles through the application of high pressure has shown great promise [22–24]. The application of high pressure (several GPa) can effectively consolidate ceramics such as alumina, silica and ferroelectrics, even at room temperature with a density >90%. In addition, these consolidates retained their nanosize grain structure and were free from secondary phase(s) from sintering additives. These studies inspired the present authors to explore the potential of a high-pressure compression technique in consolidating ultrahigh-temperature ceramics (UHTC), like TaC.

High-pressure consolidation of TaC at room temperature is expected to be deformation-dominated, considering the negligible chance of atomic diffusion. Thus, the high-pressure consolidation of TaC can possibly give new insight into the deformation mechanism leading to the manufacturing of an engineering component. The deformation behavior of TaC becomes different from other ceramics, due to the presence of a majority of Ta–Ta metallic bonds, unlike other ceramics, which have a majority of ionic and covalent bonds. The presence of a good amount of metallic bonds in TaC activates favorable slip planes and produces plastic deformation in a significant amount as compared to similar high-temperature ceramics, even at low temperatures [1,25–27]. However, these studies on room-temperature deformation of TaC are carried out using indentation based deformation and are limited to a very small volume [1,25–27]. The application of high hydrostatic compression over the entire volume of the body is expected to activate all possible deformation mechanisms to accommodate the severe plastic strain generated. Thus, it is possible to observe several possible low temperature deformation mechanisms in TaC over the large volume of high-pressure consolidate as compared to confined small volume beneath the indent.

In the context of the above discussion, the present study investigates an ultrahigh-pressure compression technique as a tool for room-temperature consolidation of TaC. The deformation mechanisms operating at room temperature and high hydrostatic pressure are analyzed with the help of high-resolution transmission electron microscopy (HR-TEM). Efforts are made to understand the consolidation mechanism of TaC at room temperature in terms of the structural changes taking place as a result of active deformation phenomena. A similar study was performed to consolidate TaC at high temperature (1830 °C) and ultrahigh pressure to understand and compare the deformation mechanisms operating at room and high temperature. The mechanical properties (elastic modulus and hardness) of TaC consolidated at RT and 1830 °C are also reported.

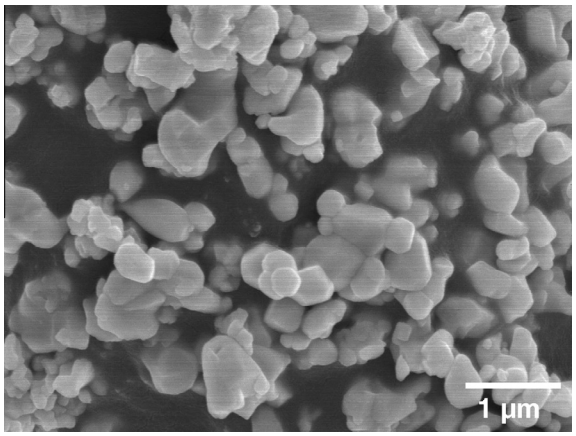

Fig. 1. SEM micrograph of the as-received TaC powder. The average particle size is 0.36 ± 0.13 μm.

2. Materials and methods

Fine tantalum carbide powder, with an average particle size of 0.36 ± 0.13 μm, was obtained from Inframat advanced Materials LLC (CT, USA). The TaC powder has a purity of 99.7% with the total carbon content $\geqslant$6.2 wt.%, free carbon $\leqslant$0.15 wt.%, Nb < 0.3 wt.% and O in the range of 0.13–0.3 wt.%. Fig. 1 shows an SEM micrograph of as-received fine TaC powders. The high-pressure (HP) consolidation of TaC was performed at 7.7 GPa at room temperature (RT) and high temperature (HT; 1830 °C). The experiments were carried out using a toroidal-type chamber (a detailed description of this high-pressure method is given elsewhere [28,29]) assembled in a 1000 t hydraulic press. The precursor powder was initially pre-compacted in a piston cylinder type up to 0.2 GPa. For the HP-RT experiments, the pre-compacted samples were placed into a lead container, which acts as a quasi-hydrostatic pressure-transmitting medium. The external diameter of the lead pressure cell is 12.0 mm, with an internal diameter of 8.0 mm and height of 12.0 mm [22]. This container was then put into a ceramic gasket that works as a sealing between the two parts of the toroidal-type chamber, to achieve the desired pressure. For the HP-HT experiments, the pressure cell consisted of a graphite heater (height of 12.0 mm, external diameter of 12.0 mm, internal diameter of 8.0 mm and wall thickness of 2.0 mm), two small discs of fired pyrophyllite and two h-BN discs (diameter of 8.0 mm). A capsule of h-BN (6.0 mm internal diameter) is placed between these discs, with the sample inside. A schematic of this container is shown in Fig. 2a. In this experimental set-up, h-BN acts as a nearly hydrostatic pressure-transmitting medium. The temperature was measured with a chromel–alumel type K thermocouple encapsulated in an Al_2O_3 sleeve. In both experiments (RT and HT), the pressure calibration was performed by the "fixed points" technique [28]. The pressure is accurate to ±0.5 GPa. A small piece of Bi was put on top of the sample in order to check the applied