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Room and ultrahigh temperature structure-mechanical property relationships of tungsten alloys formed by field assisted sintering technique (FAST)

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ABSTRACT

Tungsten based alloys have become of critical importance in a number of applications including plasma-facing materials in nuclear fusion reactors, rocket nozzles for aerospace applications, and in kinetic energy penetrators in the defense industry. Formation of components for these uses by powder metallurgical techniques has proven challenging, due to tungsten's relatively poor sinterability. Here we report the use of field assisted sintering technique (FAST) to produce high density, fine grain alloys with mechanical properties comparable or superior to that of components produced by conventional techniques. Alloys of pure tungsten, W-3 vol%TiC, W-5 vol%TiC, and W-10 vol%Ta were synthesized at 2100 °C, 35 MPa for 25 min using FAST. Microstructural characterization revealed effective reduction of grain size with TiC addition and preferential diffusion of oxygen into the center of tantalum particles in tantalum containing alloys. Tensile testing of alloys revealed TiC addition to W resulted in substantially improved ultimate tensile strength at the cost of ductility in comparison at temperatures up to 1926 °C (3500 °F) however this strengthening effect was lost at 2204 °C (4000 °F). Addition of 10 vol%Ta to W resulted in reduced hardness at room temperature, but substantially increased yield strength at the cost of slightly reduced ductility at 1926 °C and 2204 °C.

1. Introduction

Tungsten based alloys have established themselves in a number of applications ranging from plasma facing materials [1,2] in nuclear reactors to rocket nozzles [3,4] due to the naturally high hardness, elastic modulus, and melting temperature of tungsten. Unfortunately, use of tungsten in structural components has traditionally been hindered by tungsten's brittleness at room temperature, as the ductile-to-brittle transition temperature (DBTT) [5] of typical metallurgical grade tungsten is several hundred degrees Celsius higher than room temperature. Refinement of tungsten to remove interstitial elements such as oxygen and nitrogen can improve the ductility of tungsten [6,7], but purification to the levels required to impart substantial ductility is prohibitively expensive and impractical in most instances. Therefore, techniques capable of improving the mechanical properties of tungsten alloys while still allowing for economical production of tungsten components are highly desirable.

The high melting temperature of tungsten in comparison to other metals places limitations on the techniques that can be used for production of its alloys [8]. Techniques such as arc melting [9] and casting are capable of producing sufficiently high temperatures to melt tungsten alloys, however they produce materials of unacceptable grain size. Cast billets of these alloys must therefore undergo further forging and processing to obtain reasonable mechanical properties, substantially increasing the cost and processing time of these components.

Powder metallurgical techniques offer advantages over these melting techniques as they can directly produce net-shape components of these alloys at reduced temperature [8]. Components generally require reduced post-processing and machining in comparison to casting techniques, and possess a comparatively equiaxed grain structure. Unfortunately, sintering of tungsten by conventional techniques such as pressureless sintering or hot pressing frequently results in components of low density, requiring additions of other metal elements such as Fe or Ni to make use of liquid phase sintering [10,11]. While such additions

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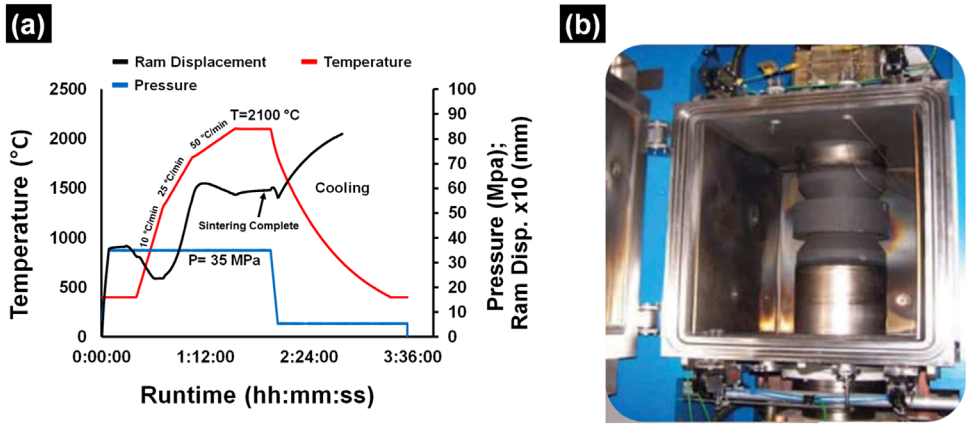

Fig. 1. (a) Example temperature, pressure, and ram displacement curves collected during sintering of W-5 vol%TiC Alloy, and (b) image of 250 ton SPS system.

substantially reduce the processing temperature of these alloys, the high concentration of non-refractory metals makes these alloys unacceptable for use in high temperature applications.

Field assisted sintering technique (FAST), also known as spark plasma sintering (SPS), is a non-conventional powder consolidation technique which uses concurrent application of pressure, temperature, and high electrical current density to sinter metallic and ceramic parts [12]. FAST has been demonstrated capable of producing high density components at a reduced sintering temperature in comparison to conventional sintering techniques for a number of material systems [13]. FAST components also display reduced grain growth due to the shorter sintering cycle time inherent in the process which is a result of the higher heating and cooling rates offered by heating via the electric field [14]. FAST is therefore a promising technique for producing industrial scale alloys of high-temperature refractory materials such as SiC [15] or tungsten [16].

FAST consolidation has been previously conducted on a number of tungsten alloys [16–18]. While sintering of pure tungsten has been studied, a large interest exists in consolidation of tungsten with additional additives to improve its mechanical properties through alloying effects or strengthening mechanisms such as precipitation hardening or grain size reduction. Of these additives, transition metal oxides such as La_2O_3 [19] and carbides such as HfC [20] are of primary interest due to their high temperature stability and generally low reactivity with tungsten. If particles of these additives are sufficiently small, they produce a Zener pinning effect which greatly impedes grain growth during sintering and improves the recrystallization resistance of the base alloy [21,22]. Additives of other refractory metals including tantalum [23] and molybdenum [24] have also been investigated due to their full miscibility with tungsten. Alloying with these elements has been shown lower the DBTT of tungsten and improve its yield strength (YS) and ultimate tensile strength (UTS). These additives have largely been incorporated into refractory materials by arc melting, with little work done investigating the high-temperature performance of these alloys produced by FAST [25].

Of tested additives, titanium carbide (TiC) is one of the most common nanoparticle additions for grain growth inhibition due to its high melting temperature and hardness as well as relatively low cost [26–28]. Generally, alloying additions are kept at a relatively low concentration to obtain desired grain refinement and second phase precipitation hardening without causing significant changes to the overall high-temperature properties of the tungsten alloy. As TiC displays little ductility even at elevated temperatures, it is also of interest to investigate W-Ta alloys who should display large ductility above the alloy's DBTT while possibly also showing increased strength due to solid solution strengthening effects [29]. In this work, we report investigations into microstructure and mechanical properties of W, W-TiC, and W-Ta alloys produced by FAST.

2. Experimental procedure

M10 grade tungsten powder was obtained from Global Tungsten Products Inc., tantalum powder was obtained from H.C. Starck, and titanium carbide powder (TiC) acquired from U.S. Research Nanomaterials for all sintering investigations. Tungsten powder was treated at 950 °C for 4 h under hydrogen prior to use in order to reduce oxide contamination. Powders were then weighed in quantities equivalent to the theoretical values needed to obtain fully dense sintered products for alloys of W-3 vol%TiC, W-5 vol%TiC, and W-10 vol%Ta. Powders were then combined in Nalgene bottles with tungsten carbide media in inert atmosphere at a ball-to-powder ratio (BPR) of 4:1: Filled bottles were placed on a rotary ball mill operating at 150 RPM and milled for four hours to evenly combine precursor powders without introducing substantial mechanical alloying or contamination to the powder blend. Powders were then sieved under inert atmosphere to remove milling media. Small quantities of powder from each alloy batch were separated for SEM and X-ray diffraction analysis. The remainder of the powder was then quickly transferred to B_4C coated graphite dies designed for production of $4'' \times 4''$ (10.16×10.16 cm) tiles. Dies were then transferred into the FAST sintering chamber and placed under vacuum. Powders were sintered at 2100 °C, 35 MPa, for 25–30 min under vacuum. An example of the measured temperature, pressure, and shrinkage curve obtained of a tungsten alloy during sintering is shown in Fig. 1.

Following sintering, pressure application was released, the tile was allowed to cool naturally to room temperature, and was then removed from the die system.

After removal of the tile from the die, tensile test samples were cut from each tile using wire EDM method. Cut bars were then sand blasted and polished by hand in order to remove residual oxide and surface damage introduced by the cutting process. Reaction of carbon from the graphite die used for sintering resulted in the formation of an outer carbide layer around the sample, therefore care was taken to ensure complete carbide removal from samples prior to polishing. The density of sintered samples was determined using the Archimedes' Principle water displacement method using these cleaned tensile bar samples. Acoustic measurements were performed on tensile bar sections to determine the Elastic modulus and Poisson's ratio of each alloy. Sections from tile sections between where tensile test bars were removed were