Microstructure and mechanical properties of mechanically alloyed and solid-state sintered tungsten heavy alloys

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Abstract

The mechanical properties of solid-state sintered 93W–5.6Ni–1.4Fe tungsten heavy alloys fabricated by mechanical alloying were investigated. Blended W, Ni and Fe powders were mechanically alloyed in a tumbler ball mill at a milling speed of 75 rpm employing a ball-to-powder ratio of 20:1 and a ball filling ratio of 15%. A nanocrystalline size of 16 nm and fine lamellar spacings of 0.2 μm were obtained in mechanically alloyed powders at a steady state milling stage. Mechanically alloyed powders were consolidated into green compacts and solid-state sintered at 1300°C for 1 h in a hydrogen atmosphere. The alloys sintered from mechanically alloyed powders showed fine tungsten particles (about 3 μm in diameter) and a relative density above 99%. The volume fraction of the matrix phase was 11% and the tungsten/tungsten contiguity was determined to be 0.74. The alloys exhibited high yield strengths (about 1100 MPa) due to their fine microstructures, but exhibited reduced elongation and impact energy due to a large area fraction of tungsten/tungsten boundaries and the low volume fraction of matrix phase. © 2000 Elsevier Science S.A. All rights reserved.

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1. Introduction

Liquid-phase sintered tungsten heavy alloys (WHAs) show a typical microstructure where bcc tungsten particles, 30–40 μm in diameter are embedded in a fcc W–Ni–Fe solid solution matrix. Generally, liquid-phase sintered WHAs are fabricated by blending raw powders of tungsten, nickel and iron and sintering at a temperature above 1460°C in a hydrogen atmosphere [1]. Due to their combination of high density, strength and ductility, WHAs are used as kinetic energy penetrators, counterweights, radiation shields, vibration damping devices, and electrical contacts [2]. Recently, numerous investigations have been carried out to improve mechanical properties of heavy alloy, including their penetration capabilities as kinetic energy penetrators [3–6]. Penetration capabilities are known to improve by enhancing the adiabatic shear deformation resulting in the self-sharpening behavior of kinetic energy penetrators [7–10]. It was reported that adiabatic shear deformation could also be enhanced by refinement of the microstructures of WHAs [11]. The mechanical properties of WHAs are known to improve through refinement of their microstructures [12]. Many researchers have investigated refined microstructures of WHAs via several techniques including alloying refractory elements such as Mo and Re [13], cold working followed by recrystallization [14], solid-state sintering [15], and mechanical alloying (MA) [16]. The MA process, developed by Benjamin [17], is an advanced fabrication process that can produce ultra-fine and homogeneous alloy powders [18]. Therefore, MA has been suggested as a process to develop advanced WHAs with improved mechanical properties [11]. Various WHAs have been mechanically alloyed using attritors, Spex mills and tumbler ball mills and then consolidated by solid-state sintering at temperatures ranging from 1200 to 1425°C [19–21].
Although earlier results showed that WHAs with the fine microstructure could be successfully processed, the limited results have presented the mechanical properties of mechanically alloyed WHAs. In this study, tensile and fracture properties of a mechanically alloyed and solid-state sintered 93W–5.6Ni–1.4Fe heavy alloy are compared with those of a liquid-phase sintered 93W–5.6Ni–1.4Fe heavy alloy.

2. Experimental procedures

Tungsten powders of 2.5 μm in diameter, nickel powders of 2.5 μm in diameter and iron powders of 3.5 μm in diameter were mechanically alloyed in a tumbler ball mill to produce WHAs with a composition of 93W–5.6Ni–1.4Fe. The diameter of the mill was 255 mm, and tool steel balls of 8 mm in diameter were used as the milling media. The milling speed was 75 rpm, milling time varied up to 72 h, ball-to-powder ratio was 20:1 by weight, and the ball-filling ratio was 15% by volume. The crystallite sizes of mechanically alloyed tungsten heavy alloy powders were measured from the broadening of X-ray diffraction (XRD) peaks, and lamellar spacing was measured using scanning electron microscopy (SEM). The mechanically alloyed powders were compacted into green compacts and sintered at 1300°C for 1 h in a hydrogen atmosphere, and mixed powders were sintered at 1485°C for 1 h. The densities of the sintered specimens were measured by the Archimedes water immersion method. The tungsten particle sizes, matrix volume fraction, and tungsten/tungsten contiguity of the sintered alloy were measured by using SEM. The sintered specimens were subsequently annealed at 1150°C for 1 h in a nitrogen atmosphere and water-quenched in order to prevent hydrogen embrittlement and impurity segregation such as P and S [22,23]. The heat-treated samples were tensile tested at a strain rate of $1.33 \times 10^{-3}$ s$^{-1}$, and impact tests were performed on unnotched samples using an instrumented Charpy impact test. The failure mechanisms of the alloys were analyzed from fractographic observations of failed tensile and impact-tested samples.

3. Results and discussion

Fig. 1 shows the microstructure of mechanically alloyed 93W–5.6Ni–1.4Fe powders with milling speed of 75 rpm, ball-to-powder ratio 20:1, ball filling ratio 15%. A powder mechanically alloyed for 1 h shows a mixture of tungsten (bright phase), nickel and iron powders (dark phase) due to the repeated welding of elemental powders taking place during MA (Fig. 1a). Fig. 1b exhibits a fine lamellar structure of mechanically alloyed powders with milling time of 48 h. Fig. 2 shows the variation of the average powder size and average lamellar spacing of mechanically alloyed powders. The average particle size increased linearly with milling time up to 16 h due to the predominance of particle welding during this period. Subsequently, the average particle size decreased as fracture of the heavily deformed mechanically alloyed powders began to dominate. The average lamellar spacing decreased with increasing milling time and was less than 0.2 μm in the steady state stage (after about 48 h of MA, Fig. 2b). Fig. 3a shows XRD patterns of the mechanically alloyed powders of 93W–5.6Ni–1.4Fe heavy alloys as they vary with the milling time. Broadening of the XRD peaks was observed and it was known to be associated with the refinement of the crystallite size during MA [24]. Fig. 3b shows the crystallite size of tungsten in mechanically alloyed powders measured from XRD peaks with increasing milling time. We used tungsten peaks from XRD of mechanically alloyed powders because peaks from other elements such as Ni and Fe are very weak due to their small amount. The crystallite size decreased
with increasing milling time, and the average crystallite size was about 16 nm in the steady state stage of the process.

The tungsten particle size and relative density of the material after sintering at 1300°C for 1 h are shown in Fig. 4. The relative density increased with milling time due to the mechanically activated sintering because the stored energy by severe cold working and homogeneous distribution of matrix phase by MA process enhanced the sintering process.

When mechanically alloyed for 72 h, solid-state sintered WHAs exhibited the high relative density (> 99%). The average size of tungsten particles in the sintered materials decreased to 3 μm as the milling time increased. These results indicate that MA followed by solid-state sintering is effective in refining the microstructure of WHAs.

Fig. 5 compares the microstructures of a solid-state sintered tungsten heavy alloy using mechanically alloyed powders with a liquid-phase sintered tungsten heavy alloy using mixed powders. The solid-state sintered alloy formed from mechanically alloyed powders exhibited much finer tungsten particles, about 3 μm in diameter, when compared with the 30–40 μm in diameter in conventional liquid-phase sintered heavy alloys. While the liquid-phase sintered tungsten heavy alloy manifests approximately spherical tungsten particles and the continuous matrix phase, the solid-state sintered tungsten heavy alloy showed interconnected tungsten particles. Tungsten/tungsten contiguity is a ratio of
tungsten/tungsten interfacial area to total interfacial area measured by the following equation [25],

\[ C_{WW} = \frac{2N_{WW}}{N_{WM} + 2N_{WW}} \]  

where \( N_{WW} \) is numbers of tungsten/tungsten interfaces and \( N_{WM} \) is the numbers of tungsten/matrix interfaces intercepted by a arbitrary line with unit length on the micrograph. The volume fraction of the matrix phase and tungsten/tungsten contiguity of solid-state sintered WHAs using mechanically alloyed powders, was measured as 11% and 0.74, compared to a matrix phase volume fraction of 17% and tungsten/tungsten contiguity of 0.35 for liquid-phase sintered WHAs of the same composition.

Tensile and impact tests were performed to characterize the mechanical properties of solid-state sintered alloys formed from mechanically alloyed powders and
Table 1
The comparison of microstructural parameters and mechanical properties of solid-state sintered tungsten heavy alloy using mechanically alloyed powders and liquid-phase sintered tungsten heavy alloy using mixed powders

<table>
<thead>
<tr>
<th>Microstructural and mechanical properties</th>
<th>Mechanical alloying and solid-state sintering</th>
<th>Mixing and liquid-phase sintering</th>
</tr>
</thead>
<tbody>
<tr>
<td>Tungsten particle size 3 μm</td>
<td>40 μm</td>
<td></td>
</tr>
<tr>
<td>Matrix volume fraction 11%</td>
<td>17%</td>
<td></td>
</tr>
<tr>
<td>W/W contiguity</td>
<td>0.74</td>
<td></td>
</tr>
<tr>
<td>Yield strength 1100 MPa</td>
<td>620 MPa</td>
<td></td>
</tr>
<tr>
<td>Elongation 0.5%</td>
<td>25%</td>
<td></td>
</tr>
<tr>
<td>Impact energy 5 J</td>
<td>120 J</td>
<td></td>
</tr>
</tbody>
</table>

Fig. 7. The fractograph of tensile tested specimens of (a) solid-state sintered tungsten heavy alloy using mechanically alloyed powders and (b) liquid-phase sintered tungsten heavy alloy using mixed powders.

Compared with those of liquid-phase sintered WHAs. Solid-state sintered WHAs using mechanically alloyed powders exhibited a high yield strength (about 1100 MPa) but a low elongation of 0.5% in tensile tests as shown in Fig. 6a. Liquid-phase sintered alloys of the same composition have a yield strength of 620 MPa and an elongation of 25%. Charpy impact tests of solid-state sintered WHAs using mechanically alloyed powders resulted in a low impact energy of 5 J compared with that of 120 J for liquid-phase sintered WHAs using mixed powders as shown in Fig. 6b.

The resulting improved yield strength is attributed to the refinement of the microstructural scale in the solid-state sintered WHAs prepared from mechanically alloyed powders. Ashby [25] reported that the yield strength of two-phase alloy is related to the inverse square root of average thickness of soft phase between hard phase because deformation of two-phase alloy begins with deformation of the soft phase as shown in Eq. (2)

\[ \sigma_y = \sigma_0 + k_1 G b \lambda^{-1/2} \]  

where \( \sigma_y \) is the yield strength, \( \sigma_0 \) is the intrinsic strength, \( k_1 \) is a constant, \( G \) is the shear modulus, \( b \) is the Burgers vector, and \( \lambda \) is an average thickness of matrix phase. Average thickness of matrix phase can be formulated in terms of tungsten particle size and matrix volume fraction using geometrical relationship as follows,

\[ \lambda = \frac{2}{3} D \left( \frac{V_M}{1-V_M} \right) \]  

where \( D \) is the diameter of tungsten particles and \( V_M \) is the matrix volume fraction. The relationship between yield strength and microstructural parameters, i.e. tungsten particle size and matrix volume fraction, is expressed as

\[ \sigma_y = \sigma_0 + k_2 G b \left( \frac{1-V_M}{D V_M} \right)^{1/2} \]  

where \( k_2 \) is a constant. According to Eq. (4), the yield strength of WHA increases with the decreasing tungsten particle size and matrix volume fraction. The high yield strength of solid-state sintered WHAs from mechanically alloyed powders is due to the small tungsten particle size of 3 μm and small matrix volume fraction of 11% as shown in Table 1.

Fig. 7a is a fractograph of a tensile tested specimen of solid-state sintered WHAs, and it can be compared to a fractograph of liquid-phase sintered WHA (Fig. 7b). WHAs show four fracture modes such as tungsten cleavage, matrix rupture, tungsten/tungsten interfacial debonding, and tungsten/matrix interfacial debonding. Churn and Yoon [26] reported that cracks form preferentially at the tungsten/tungsten interfacial boundaries and link up to cause catastrophic failure because those boundaries are the weakest. Most fractured interfaces are composed of tungsten/tungsten interfacial boundaries in solid-state sintered WHAs using mechanically alloyed powders, while tungsten cleavage and matrix rupture are mainly shown in liquid-phase sin-
tered WHAs using mixed powders. The brittle fracture behavior of solid-state sintered tungsten heavy alloy is attributed to the high tungsten/tungsten contiguity and low matrix volume fraction. Higher tungsten/tungsten contiguity provides preferential sites for microcrack nucleation, and lower matrix volume fraction provides reader microcrack link up [27].

The comparison of microstructural parameters and mechanical properties was made between a 93W–5.6Ni–1.4Fe heavy alloy made by solid-state sintering of mechanically alloyed powders, and one made by conventional liquid-phase sintering of mixed powders as listed in Table 1. Table 1 reveals the tensile and fracture behavior of mechanically alloyed WHAs are strongly affected by the microstructural parameters such as tungsten particle size, matrix volume fraction and tungsten/tungsten contiguities.

4. Conclusions

The mechanical properties of WHAs, fabricated using mechanically alloyed powders and employing solid-state sintering, were investigated. When solid-state sintered at 1300°C, WHAs showed ultra-fine tungsten particles of about 3 μm with high density above 99%. These solid-state sintered WHAs using mechanically alloyed powders exhibited a high yield strength of about 1100 MPa due to a fine microstructure, but showed reduced elongation and impact energy due to a large area fraction of brittle tungsten/tungsten interfaces and low matrix volume fraction.

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