EVALUATION OF MICROSTRUCTURAL INHOMOGENEITY IN LIQUID PHASE SINTERED TUNGSTEN HEAVY ALLOYS

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ABSTRACT

In order to study microstructural inhomogeneity of tungsten heavy alloys, 78W-15.4Ni-6.6Fe tungsten heavy alloys that were liquid phase sintered under both ground-based and microgravity conditions were investigated. Standard metallographic procedures were used to obtain optical micrographs. Microstructural inhomogeneity was observed in both ground based and microgravity samples. Parameters, such as solid volume fraction, contiguity, connectivity and area equivalent grain size increased from the top to the bottom in the ground-based sample, and increased from the periphery to the center in the microgravity sample. A digital image analysis approach called tessellation was employed to measure the liquid phase area distributions. Histograms of the measured liquid phase areas were plotted. 3D computer simulation was conducted to obtain random microstructures. Comparing with the random microstructures, liquid phase area distributions were found inhomogeneous in both ground based and microgravity samples.

INTRODUCTION

Liquid phase sintering is a widely used method for net shaping from powders [1]. The liquid provides a capillary force that pulls the solid grains together and offers a mass transport medium. Rearrangement of the solid grains takes place once the liquid forms. Then solution-reprecipitation occurs. In this process, small grains dissolve into liquid and reprecipitate on the large grains. Rearrangement and solution-reprecipitation result in dense packing of the solid grains. Thus, a liquid aids in densification during sintering.

One common concern about liquid phase sintering is the segregation of the solid grains from the liquid. This is due to the density difference between solid grains and the liquid, which causes the solid to settle toward the bottom. This is more pronounced in tungsten heavy alloys (W-Ni-Fe or W-Ni-Cu) since the density difference between tungsten grains and the liquid is large (near 9 g/cm³). Many works have reported the segregation of tungsten grains from the liquid along the
gravitational direction, resulting in higher solid content on the bottom than on the top [2-5]. Practically, liquid phase sintering of tungsten heavy alloys is restricted to high solid content compositions; however, microstructural segregation is still evident even with low liquid content alloys. In microgravity, there is no gravity-induced microstructural segregation, but microstructural segregation has still been observed [6, 7]. The compact was spheroidized and solid content increased toward the center of the compact. A high liquid content region formed at the periphery.

This work analyzed the microstructural inhomogeneity in both ground based and microgravity samples, including solid volume fraction, contiguity, connectivity, area equivalent grain size, mean intercept and liquid phase area distribution. A computer simulation program was employed to reconstruct the random microstructures. The experimental microstructures were compared with simulated ones to observe the inhomogeneity of liquid phase area distributions.

EXPERIMENTAL PROCEDURES

Elemental powders of W, Ni, and Fe were mixed according to the composition of 78 wt% W-15.4 wt% Ni-6.6 wt% Fe and cold isostatically pressed into cylinders. The green compacts were presintered at 1400°C in a vacuum furnace for 1 hour and then dry machined into cylinders of 10 mm in diameter and 10 mm in height. These samples were then placed in individual alumina crucibles and vacuum-sealed in BN cartridge tubes. The vacuum-sealed samples were liquid phase sintered at 1500°C for 120 min on ground base and in microgravity of the space shuttle Columbia (MSL-1 flight, 1997). In both cases, the heating rate was 10 °C/min. More details have been described elsewhere about the preparation of the samples and the sintering procedure [8].

MICROSTRUCTURE ANALYSIS

After sintering, all samples were sectioned along the gravitational direction during pre-sintering, then mounted and polished to 0.05 μm finish for metallographic analysis. An image analysis system was employed to measure solid volume fraction and area equivalent grain size. Contiguity and connectivity were measured manually. The liquid phase area distribution was obtained using tessellation approach. The gray scale micrographic images were first processed into binary images by a sequence of operations, including binary thresholding, hole filling, grain segmentation, and erosion/dilation. After erosion/dilation, the grains were separated from each other. Then the tessellation operation generated cells by iteratively expanding each grain simultaneously. A network of the expanded grain boundaries was obtained. The segmented grain structure was superimposed into the network. The area within each cell on the network was known as the feature area that was measured by feature analysis. In order to accommodate the grain size effects on the measured cell area, the grain area was substituted from each cell area. Then the reported cell area was the difference in the area of the tessellated cell and the area of the grain that caused the cell. The tessellation results were plotted as frequency vs. cell area. The accumulative frequencies were also plotted.

The microstructures of the sintered samples were compared with the random microstructures. A computer simulation program was employed to construct some random microstructures of 78W-15.4Ni-6.6Fe alloy. The details of this program have been reported elsewhere [9]. This program took into account gravity effect, dihedral angle, solid volume fraction and grain size distribution which were obtained from the microstructures of the sintered samples. The simulation
conditions corresponded to ground based and microgravity sintering which was at 1500°C for 120 min. The computer program first constructed some three-dimensional microstructures, and then it cut the three-dimensional microstructures along a random plane to obtain two-dimensional representatives as usually encountered in a metallographic section. After the two-dimensional microstructures came out, the tessellation approach was employed as with the microstructures of the sintered samples.

DEFINITION OF INHOMOGENEITY IN TESSELLATION

Tungsten heavy alloys have a microstructure consisting of tungsten grains and solidified liquid matrix. Ideally, if the tungsten grains are mono-sized and liquid matrix is uniformly distributed around tungsten grains, the measured liquid phase areas in the tessellation operation will have only a single value. In practice, tungsten grains are not mono-sized and agglomerate together during sintering. The liquid matrix around the tungsten grains is also not uniformly distributed and has a distribution of areas. Computer simulation took into account the grain size effects and generated microstructures which were assumed to be homogeneous compared with those of sintered samples. In tessellation operation, the narrower the liquid area distribution, the less the microstructural inhomogeneity.

RESULTS AND DISCUSSIONS

Photographs of ground based and microgravity sintered samples after sectioning are showed in Figure 1. The difference in the samples is obvious. The ground base sintered sample distorted. Because of the gravity effect, tungsten settled to the bottom, leaving a liquid dome on the top. The microgravity sample, however, had a spheroidized shape. No obvious liquid dome was found on the microgravity sample.

![Figure 1](image-url)

(a) (b)

Figure 1. Photos of 78W-15.4Ni-6.6Fe alloys after sintering at 1500°C for 120 min under (a) ground-base and (b) microgravity conditions.

Figures 2 and 3 show the microstructures of the ground-base and microgravity sintered samples, respectively. Table I gives their corresponding measured microstructural parameters, including solid volume fraction, contiguity, connectivity and area equivalent grain diameter. The microstructures were not homogenous. For the ground based sample, tungsten volume fraction increased from 39.3 vol.% at the top to 74.3 vol.% at the bottom. Contiguity, connectivity and area equivalent grain size all increased from the top to the bottom. These reflect the gravitational effects on microstructural inhomogeneity in liquid phase sintering on earth. Gravity induces
settling of tungsten grains, resulting in high tungsten content in the bottom. As tungsten content increases, contiguity, connectivity and grain size also increase. For the microgravity sample, tungsten volume fraction increased from 52.4 vol.% at the periphery to 68.5% at the center. Contiguity, connectivity and area equivalent grain size also increased from the periphery to the center. The microstructure inhomogeneity of microgravity sample was caused by the surface tension that minimizes the total energy. However, the microstructures of the microgravity sample were not so inhomogeneous as those of the ground based sample.

![Microstructures](image)

(a) ![Microstructures](image)

(b) ![Microstructures](image)

(c) ![Microstructures](image)

Figure 2. Microstructures of 78W-15.4Ni-6.6Fe alloy after sintering at 1500°C for 120 min in ground base: (a) top, (b) middle and (c) bottom.
Figure 3. Microstructures of 78W-15.4Ni-6.6Fe alloy after sintering at 1500°C for 120 min in microgravity: (a) periphery and (b) center.

Table I. Measured microstructures of 78W-15.4Ni-6.6Fe alloys after sintering at 1500°C for 120 min in ground base and microgravity (Parenthesis is the standard deviation).

<table>
<thead>
<tr>
<th>Microstructure</th>
<th>Ground based</th>
<th>Microgravity</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Top</td>
<td>Middle</td>
</tr>
<tr>
<td>Solid volume fraction (vol.%)</td>
<td>39.31</td>
<td>56.64</td>
</tr>
<tr>
<td></td>
<td>(9.58)</td>
<td>(6.26)</td>
</tr>
<tr>
<td>Contiguity</td>
<td>0.086</td>
<td>0.141</td>
</tr>
<tr>
<td></td>
<td>(0.011)</td>
<td>(0.038)</td>
</tr>
<tr>
<td>Connectivity</td>
<td>0.757</td>
<td>0.901</td>
</tr>
<tr>
<td></td>
<td>(0.128)</td>
<td>(0.109)</td>
</tr>
<tr>
<td>Area equivalent grain diameter (μm)</td>
<td>43.77</td>
<td>45.38</td>
</tr>
<tr>
<td></td>
<td>(2.94)</td>
<td>(1.84)</td>
</tr>
</tbody>
</table>

Figure 4 shows the microstructure in the middle of the ground base sintered sample after tessellation. Figure 5 shows the histograms of the tessellated cell areas of the ground base sintered sample. Figure 5 (a) has the broadest distribution and a model of cell area at 1600 μm, while Figure 5 (c) has the narrowest distribution and a model of cell area at 800 μm. These results indicate that liquid area distribution was not homogeneous. It was more inhomogenous at the top than at the bottom. Liquid distribution of the microgravity sintered sample was more inhomogenous in the periphery than at the center as shown in Figure 6. Figure 6 (a) has broader distribution then Figure 6 (b). The model of the cell areas in Figure 6 (a) is also bigger than that in Figure 6 (b).
Figure 4. Tessellated microstructure in the middle of the sample after sintering at 1500°C for 30 min in ground base.
Figure 5. Histograms of cell areas of experimental 78W-15.4Ni-6.6Fe alloy after sintered at 1500°C for 120 min in ground base: (a) top, (b) middle and (c) bottom.
Figure 6. Histograms of cell areas of experimental 78W-15.4Ni-6.6Fe alloy after sintered at 1500°C for 120 min in microgravity: (a) periphery and (b) center.
Figure 7(a) gives an example of the computer simulated random microstructure which corresponded to the middle of the ground base sintered sample. Figure 7(b) is the corresponding microstructure after tessellation. Tungsten grains in the random microstructure were spherical which was assumed by the computer simulation. Sinter bonds between tungsten grains had formed. The clusters of tungsten grains were also present. Figure 8 shows the histograms of the liquid areas of the random microstructures corresponding to ground base. Due to high solid volume fraction, computer could not generate the random microstructure corresponding to the bottom of the ground base sintered sample. Figure 9 shows the histograms of the liquid areas of the random microstructures corresponding to microgravity. The random microstructures had narrower distributions of liquid areas than those of the experimentally sintered samples. These indicate the inhomogenous distributions of the liquid phase in the sintered samples.

Figure 7. Random microstructures corresponding to the middle of the sample after sintering at 1500°C for 30 min in ground base: (a) original and (b) tessellated.
Figure 8. Histograms of cell areas of simulated microstructures of 78W-15.4Ni-6.6Fe alloy after sintered at 1500°C for 120 min in ground base: (a) top and (b) middle.
Figure 9. Histograms of cell areas of simulated microstructures of 78W-15.4Ni-6.6Fe alloy after sintered at 1500°C for 120 min under microgravity: (a) periphery and (b) center.
It was observed that agglomeration of tungsten grains occurred even under microgravity. It is a natural event. According to the observation of Tewari et al., no isolated grains were present in both ground base and microgravity sintered tungsten heavy alloys (10). The agglomeration effect is the result of energy minimization for the microstructure (11). The solid contact between two tungsten grains is stable because the total solid-solid surface energy is less than the two solid-liquid surfaces that are replaced by the solid contact. The consequence of grain agglomeration is the inhomogeneous distribution of the liquid in the sintered compact.

CONCLUSIONS

The microstructures of both ground base and microgravity liquid phase sintered tungsten heavy alloys were inhomogeneous. For ground base sintered sample, the solid volume fraction, contiguity, connectivity and grain size increased from the top to the bottom. For the microgravity sample, these parameters increased from the periphery to the center. The tessellation approach showed that liquid distributions were not uniform in the microstructures. In ground base, the inhomogeneity of the liquid distributions decreased from the top to the bottom, while the inhomogeneity of liquid distributions decreased from the periphery to the center in microgravity. Compared with the simulated random microstructures, microstructures of experimentally sintered samples were more inhomogeneous.

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REFERENCES


