Sintering Characteristics of Zr-U Pellets in High-vacuum Environment

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1. Introduction

Zr-U alloys have been considered as one of candidate nuclear fuels for fast reactors due to their excellent thermal conductivity [1-3]. They have occasionally been fabricated by powder metallurgy processes [4]. The powder metallurgy is usually composed of three processes; blending, compacting and sintering. During sintering of Zr-U pellets at high temperatures, the high-vacuum environment is usually applied to avoid undesirable oxidation reaction. In this case, it was observed that the vapor phenomena of U in the surface of sintered Zr-U alloy were apparent when the sintering temperatures over than 1300°C were applied [5]. This means that the sintered Zr-U alloys would show a difference in the concentration of alloying elements in their surface and center parts, indicating the reduction in the homogeneity of alloying elements. In this study, the distribution of alloying elements of Zr-U alloy sintered at 1450°C for 3 hours in the high-vacuum of 10^-6 torr were evaluated.

2. Methods and Results

2.1 Sintering of Zr-U pellets

The Zr-powders were prepared by a hydriding-dehydriding process. Their impurities were analyzed to be about (in ppm) 3040 O, 80 C, 50 N and 50 H. The U-powders were prepared by an atomizing process, and their impurities were analyzed to be about (in ppm) 2500 O, 220 C, 100 N and 10 H. The Zr and 38 wt.% U powders were mixed and then compacted to form cylindrical-type pellets with 25 mm in diameter and 35 mm in height. The compacted Zr-U pellets were put in the crucible placed in the vacuum furnace for sintering. The sintering of pellets was performed at 1450°C for 3 hours under a high-vacuum environment.

During sintering, the products deposited in the inner surface of crucible were examined using SEM/EDS. Their crystal structures were observed using a XRD. The sintered Zr-U alloys were subject to prepare specimen in two zones (surface and center) for the examination of chemical composition.

2.2 Vaporization behavior of U during sintering

Figure 1 shows SEM/EDS results of film deposited at the surface of crucible during sintering at 1450°C for 3 hours. The deposited film mainly consisted of (wt.%) 87.6U, 8.2O and 4.2Zr. This means that the metallic U would be possible to vapor in these sintering conditions. The equilibrium vapor pressure of pure U is possible to know as well known formula; \[ \log P \text{ (atm)} = -\frac{26,210 \pm 270}{T} + (5.902 \pm 0.135) \]. Theoretical temperature and vacuum degree for the vapor of U could be possible to explain the vapor of U during sintering. With regard to the Zr component in deposited film, it is believed that the some of Zr would participate in the vapor of U, because the equilibrium phase is a single gamma-Zr.

<table>
<thead>
<tr>
<th>Element</th>
<th>Wt.%</th>
<th>At.%</th>
</tr>
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<tbody>
<tr>
<td>U</td>
<td>87.6</td>
<td>39.8</td>
</tr>
<tr>
<td>Zr</td>
<td>4.2</td>
<td>5.0</td>
</tr>
<tr>
<td>O</td>
<td>8.2</td>
<td>55.2</td>
</tr>
</tbody>
</table>

Figure 2 shows the X-ray diffraction pattern on the deposited film. The phase of deposited film was analyzed to be a UO₂ (cubic, a=0.5466 nm). During sintering, the vacuum should be done around 10^-6 torr to avoid the oxidation of Zr-U pellets. This observation indicates that the vaporized U component would react with oxygen to form uranium oxide, even the oxygen potential is extremely low in high-vacuum environment.
2.3 Distribution of alloying elements

Figure 3 shows the chemical compositions of powder, surface and center zones of sintered Zr-U alloy. Compared with the concentrations of powder, the surface of sintered alloy showed low U and high Zr concentrations whereas the center zone exhibited similar concentrations of powders.

Figure 4 shows the concentrations of impurities of powder, surface and center zones of sintered Zr-U alloy. The sintered alloy contained much higher concentration of O than that of O in powders. This indicates that the sintering provide the oxidation reaction even though in high-vacuum environment. With regard to C and N, there was little change after sintering. However, the concentration of H significantly reduced after sintering. It would be attributed to the dehydriding reaction during sintering in high-vacuum environment.

3. Conclusion

During sintering of Zr-U pellets, the vaporization phenomena of U were observed when the sintering temperature of 1450°C and vacuum degree about 10⁻⁶ torr were applied. As the results, outer surface of sintered alloy contained high Zr and low U concentrations. In addition, the sintering in high-vacuum environment provides the increase in the O concentration whereas reduce the H concentration. These results would be useful to predict the properties of nuclear fuels.

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REFERENCES