Measurement of Thermal Expansion of Tritium Storage Alloys

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ABSTRACT

The thermal expansion property measurement was performed on the same specimens of U-Th-Zr alloys, both hydrogenated and unhydrogenated, by using a high temperature dilatometer. The determination of thermal expansion (%) over elevated temperatures and differences in the thermal behaviors among the specimens are presented.

1. INTRODUCTION

U-Th mixed fuel has been investigated mainly on oxides, carbides, molten salts and alloys for many years because of the great advantages in thorium fuel cycle system such as abundance of thorium resources, production of less transuranium elements and applicability to thermal breeder reactors. Also, mixed hydrides seem to be useful as such a fuel because both U and Th form stable hydrides. It has been theoretically indicated that MA-containing hydride fuel, U-MA-Zr-Hx, can be an appropriate target for burning MA nuclides effectively. On the other hand, several zirconium alloys have been proposed for tritium removal or storage and have been tested in different atmospheres.

Previous work by the authors showed that hydrides of U-Th-Zr alloys can be candidate materials for U-Th mixed hydride fuel. The microstructures of the alloy hydrides were very fine, where α-U phase was dispersed in the bulk of $ThZr_2H_{7-x}$ and ZrH_{2-y} phases, similar to TRIGA reactor fuel where U phase is finely dispersed in the ZrH_{2-x} bulk. They would show an inherent safety for a nuclear reactor since hydrogen atoms act not only as the moderator but also as the accelerator of neutron. For this it would be at least necessary that the hydride phases can hold hydrogen tight at elevated temperatures. Such a hydrogen holding property was observed on the U-Th-Zr alloy hydride previously.

In this paper, thermal properties of the hydrogenated U-Th-Zr alloys in terms of thermal expansion was studied in order to clarify the stability of hydride phases formed in the alloys in the high temperature region. And these results may provide a useful information on tritium handling in a nuclear fusion reactor.

2. EXPERIMENTAL AND RESULTS

The U-Th-Zr alloys with atomic ratios of U:Th:Zr = 2:1:6, 1:1:4, 1:2:6 and 1:4:10 were prepared by using an arc melting furnace and the U-Th-Zr-H with the atomic ratios of U:Th:Zr:H = 2:1:6:13.3, 1:1:4:9.5, 1:2:6:15.2 and 1:4:10:27 were prepared by using hydrogenation system. The sample dimension was about 4 x 4 x 2~3 mm^3. Thermal expansion measurements were carried out with a dilatometer. Before measurements the surface of the specimens was polished with emery papers to minimize the roughness of the surface of the specimens. Initial thickness of the specimens was measured with a digital micrometer with the accuracy of about 0.001 mm. The change in length per unit starting length is recorded as a function of temperature. The thermal expansion is obtained from the slope between two temperatures, which is defined as:

$$\text{Thermal Expansion (\%)} = \frac{\Delta L}{L_0} \times 100 \qquad (1)$$

where L_0 is the initial length of the sample at room temperature (in this experiment L_0 at 293 K), $\Delta L = L_1 - L_0$, L_1 is the lengths at temperature T_1 , respectively.

Figure 1 shows the linear expansion rates of four kinds of unhydrogenated alloys, presented together with that of UO_2 recommendation values and the NBS standard values for pure uranium, thorium and zirconium. It is shown that the expansion rates of those alloys are almost in accordance with that of zirconium. The fact that the major parts of the unhydrogenated alloys were constituted of zirconium appears to have played a decisive role in the bulk expansion of these U-Th-Zr alloys. Among the four alloys examined, three of them showed that the higher the U/Th ratio of the alloy is, the larger the expansion of the alloy becomes, though the difference in those of the alloys were considerably small. These results seem to be attributed to the fact that uranium metal has a higher thermal expansion than those of thorium and zirconium metals. The highest expansion at elevated temperature is exhibited by U_2ThZr_6, which is followed by UTh_2Zr_6, UTh_4Zr_{10} and $UThZr_4$ in decreasing order.

Figure 2 shows the linear expansion rates of four kinds of hydrogenated U-Th-Zr alloys. Similar to the unhydrogenated alloys, the hydrogenated specimens showed the same tendency, i.e. the higher the U/Th ratios, the larger the thermal expansion of the specimen becomes, with an exception of $UTh_4Zr_{10}H_{27}$, which has the highest hydrogen content with 1.93 in H/(Th+Zr) ratio. In general, hydrogenation appears to increase the thermal expansion of the U-Th-Zr alloys.

The observed increase of the thermal expansion with hydrogenation can be explained as follows. The thermal expansion is resulted from the attractive and repulsive forces between atoms in the specimen as the temperature of the specimen is increased. As the temperature rises, the energy of the specimen increases, so that the atoms vibrate to a greater degree within a short distance about a mean position. Since the repulsion term between atoms changes more rapidly than the attractive term, the potential well is not symmetrical. Thus, for a given energy or temperature, the atoms can move farther apart more readily than they can be pushed together. In case of hydrogenated specimens, hydrogen atoms move faster than other constituent atoms, which results in increase of the thermal expansion of the specimens.

In summary, hydrogenation increases the thermal expansion values of the unhydrogenated alloys. Such an experience can be applied to the tritium storage materials.

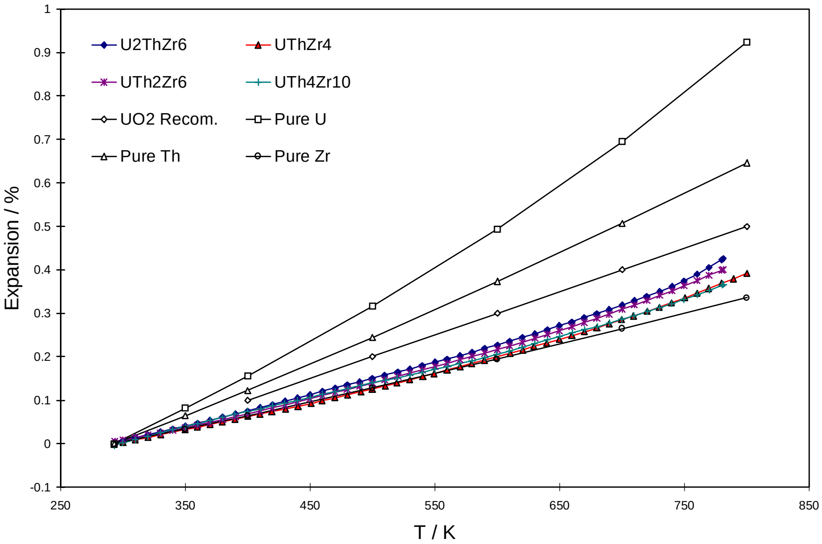

Figure 1 Thermal expansion of U-Th-Zr alloys.

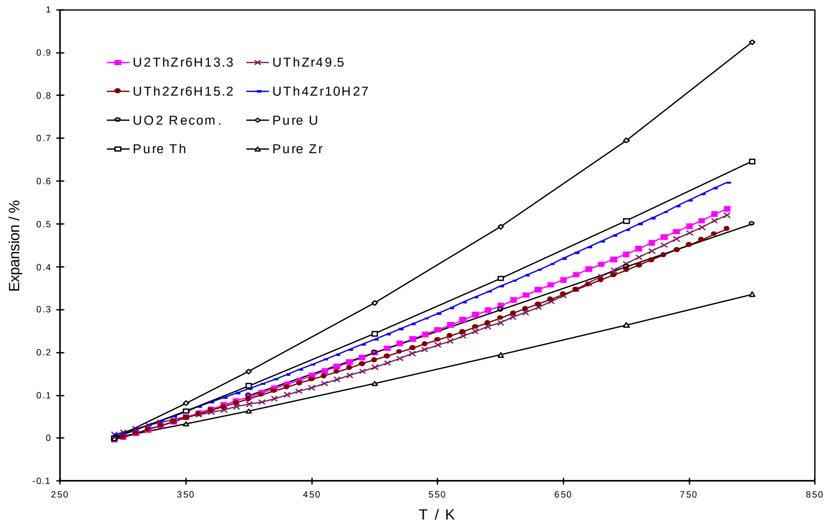

Figure 2. Thermal expansion of U-Th-Zr-H alloys.