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A Study on the Pressure-Composition Isotherm for the Reaction of ZrCo with Hydrogen

Sunmi Kim†,a Seungwoo Paek,a
Minsoo Lee,a Si-Hyung Kim,a Kwang-Rag Kim,a Do-Hee Ahn,a
Soon-Hwan Sohn,b Kyu-Min Song,b
a Korea Atomic Energy Research Institute, 150 Deokjin-Dong, Yuseong-Gu, Daejeon, 305-353 Korea
b Korea Electric Power Research Institute, 103-16 Munji-Dong, Yuseong-Gu, Daejeon, 305-380 Korea
†kimsunmi@ kaeri.re.kr

1. Introduction

The intermetallic compound of ZrCo which is one of the promising getters for the handling, transport, and storage of tritium has been extensively studied and widely used due to its attractive properties as a tritium getter. [1] At a typical storage temperature of room temperature, the ZrCo-H system has an equilibrium pressure of about 10^{-3} Pa for an absorption. The immobilized gas can be recovered at a moderate temperature. Also, from the point of view of safety, the ZrCo and its hydrides have proven to be much less pyrophoric than uranium and its hydrides are the most widely used as a tritium getter.

In the current study, the pressure-composition isotherm for the reaction of ZrCo with hydrogen was derived to evaluate its absorption and release characteristics. This provides useful information on the stability and the storage capabilities of ZrCo and the subsequent release of the hydrogen on a heating.

2. Experimental result and discussion

In this section the experimental apparatus, process of the preparation of the ZrCo fine powder, and the pressure-composition isotherm experiment are described and discussed.

2.1. Preparation of the ZrCo fine powder

The intermetallic compound of ZrCo supplied by SAES Getters (Milano, Italy) was used in the experiment.

Figure 1. Experimental apparatus

The used experimental apparatus is shown in figure 1. To powderise the ZrCo compound before the pressure-composition isotherm experiment, the ZrCo compound was weighed and taken into the reaction cell. And activation of the ZrCo compound was carried out at 773 K for 5h. Then a highly pure hydrogen gas at a pressure of about 900 torr was slowly introduced into the reaction cell. This process of an absorption and a decomposition of the hydride was repeated several times. Finally, the ZrCo compound was decrepitated into a powder upon a hydrogen sorption.

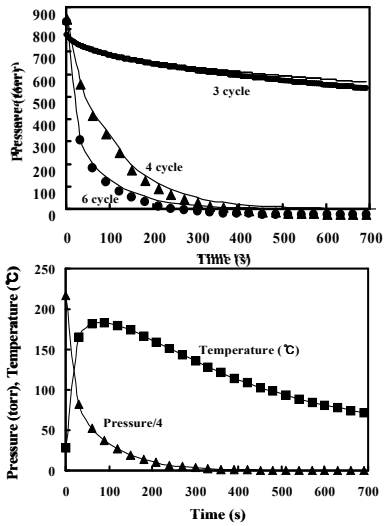

Figure 2. Above: rate variation according to the cycle of absorption and decomposition of the hydride, Below: temperature variation during the absorption and decomposition of the hydride

Figure 2 shows the rate variation and temperature variation according to the cycle of an absorption and a decomposition of the hydride. As a result, a repeated cycle of an absorption and a decomposition of the hydride led to a more rapid rate of an absorption. It was confirmed that the reaction of ZrCo with hydrogen is exothermic.

2.2 The pressure-composition isotherm

Generally, the ZrCo absorbs hydrogen isotopes down to about 10^{-4} Torr at room temperature and is heated to approximately 673 K to desorb hydrogen at around an atmospheric pressure. A highly activated free-flowing powder was used to derive the pressure-composition

isotherm for the reaction of ZrCo with hydrogen. The gas was slowly released into the reaction cell. The final pressure was recorded after an equilibrium was established. The amount of hydrogen absorbed can be calculated using ideal gas laws.

Figure 3 shows the pressure-composition isotherms for the reaction of ZrCo with hydrogen at 473 K, 523 K, and 573 K. The limiting composition for the storage of hydrogen at 473 K as read from the figure 3 is $ZrCoH_{2.8}$. The pressure-composition isotherms indicate that the length of the plateau region reduces with a rise in the temperature with a corresponding increase in the slope.

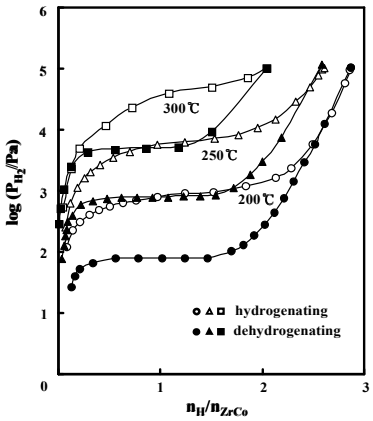

Figure 3. Pressure-composition isotherms for the reaction of ZrCo with hydrogen at 473 K, 523 K, and 573 K

2.3. The van't Hoff plot

An equilibrium plateau pressure at a given temperature is a measure of the stability of the hydride. [2] Plot of the pressure derived from the plateau region of the previous isotherm data against the reciprocal of the temperature is shown in the figure 4.

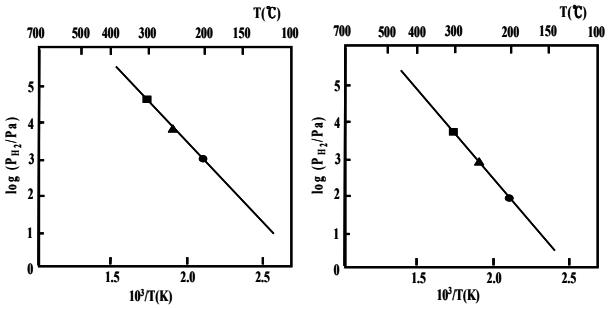

Figure 4. Left: Plot of pressure against reciprocal of temperature for the absorption, Right: Plot of pressure against reciprocal of temperature for the desorption

In the plateau region, the temperature dependency of the equilibrium pressure for a given metal/hydrogen composition can be described by the van't Hoff equation:

$$\log P = -A/T(K) + B, \quad (1)$$

in which A and B are empirical parameters of a system.
for an absorption:

$$\log P_{H2}\,(Pa) = -\,4458 / T(K) + 9.06$$

for the desorption:

$$\log P_{H2}\,(Pa) = -\,3726 / T(K) + 8.67$$

3. Conclusion

To obtain useful information on the stability and the storage capabilities of ZrCo and a subsequent release of the hydrogen on a heating, a pressure-composition isotherm experiment for the reaction of ZrCo with hydrogen at various temperatures was conducted. Thus, through the pressure-composition isotherm, we confirmed that the length of the plateau region reduces with a rise in the temperature with a corresponding increase in the slope. In addition, in the plateau region, the temperature dependency of the equilibrium pressure for the absorption-desorption of ZrCo was described by the van't Hoff equation.

REFERENCES

[1] R.-D. Penzhorn, M. Devillers and M. Sirch, Evaluation of ZrCo and other getters for tritium handling and storage, J. N. M., 170, 217-231, 1990.
[2] Yeshwant Naik, G.A. Rama Rao, V. Venugopal, Zirconium-cobalt intermetallic compound for storage and recovery of hydrogen isotopes, Intermetallics, 9, 309-312.