Measurement of ruthenium activity in Ru-Pd binary alloy

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Abstract: In order to improve the existing Ru release model from fuel in a severe accident, Ru activity (a_{Ru}) of Ru-Pd alloys was measured by a new experimental method using thermogravimetric analysis (TGA). The measured a_{Ru} agrees well with that calculated using the thermodynamic data, which verifies the accuracy of this new method.
Keywords: Ru-Pd, Thermogravimetric analysis, Ruthenium activity

1. Introduction

The latest research revealed that the Ru release rate from a nuclear fuel is affected by its activity in the Ru-Pd-Rh-Tc alloy precipitates under a severe accident [1]. This effect has not been incorporated in the existing Ru release models. On the other hand, the a_{Ru} of Ru-containing alloys is difficult to be measured by the traditional methods, such as Knudsen cell mass spectrometry, due to the low vapor pressures of Ru metal. Under this background, for improving the existing Ru release models, a new method using TGA for determining the a_{Ru} in above alloy precipitates has been proposed recently [2]. The objective of this study is to verify the accuracy and reliability of this new method by measuring the a_{Ru} in the Ru-Pd system and comparing the measured values with those calculated using the thermodynamic data in literatures [3, 4]. It is known that the Ru-Pd phase diagram calculated using the above referenced thermodynamic data perfectly agrees with that obtained by experiments [4], reflecting the high reliability of these fundamental data.

2. Experimental

The cornerstone of this new measurement method is that at one temperature (1473~1673 K) and under one Ar-(1.0~4.5 mol%) O_2 atmosphere, the oxidative vaporization rate of Ru from Ru-containing alloy powders during TGA is proportional to its a_{Ru} [2]. Therefore, the a_{Ru} of one Ru-Pd alloy should equal the ratio of the oxidative vaporization rate of Ru from this alloy powders to that from the metallic Ru powders (whose a_{Ru} equals unity) under the same experimental conditions. Based on this, the experimental procedures have been given as follows.

The binary alloy powders with the composition of $Ru_{0.5}Pd_{0.5}$ in mole fraction were prepared by a powder metallurgy method. The metallic Ru and alloy powder samples were subjected to the TGA. During the temperature rising process, Ar-5% H_2 was introduced to avoid the oxidation of samples. After reaching the equilibrium condition with no change in weight at predetermined temperatures (1523~1673 K), the atmosphere was changed to Ar-2.0% O_2. The sample weight was continuously monitored during the experiments to calculate the oxidative vaporization rate of Ru.

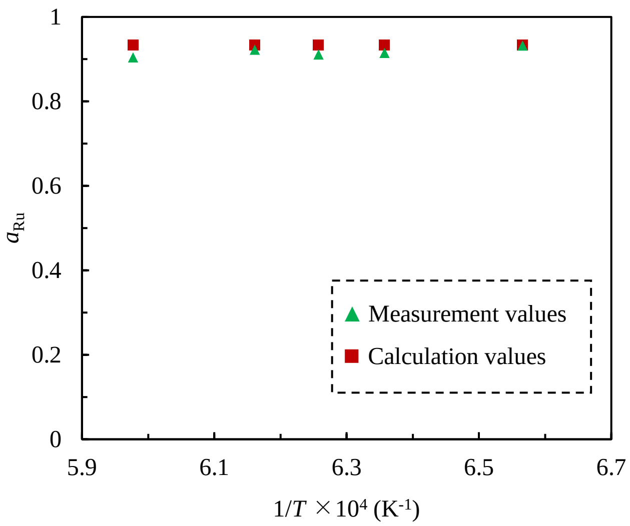

Fig. 1. Dependence of a_{Ru} on temperature for the $Ru_{0.5}Pd_{0.5}$ system.

The theoretical calculation of a_{Ru} was done by using the Thermo-Calc software, in which a self-made database containing the published thermodynamic data, such as the lattice stability values of Ru and Pd elements [3] and their interaction parameters [4], had been defined and constructed.

3. Results and discussion

Fig. 1 compares the values of a_{Ru} for the $Ru_{0.5}Pd_{0.5}$ system in the temperature range from 1523 to 1673 K measured in present study with those calculated by using the Thermo-Calc software. We can clearly see that the values of a_{Ru} measured in this study agree well with the calculated ones, indicating that the proposed new measurement method has a high accuracy.

4. Conclusions

The values of a_{Ru} for the $Ru_{0.5}Pd_{0.5}$ system measured by the new experimental method using TGA agree well with those calculated using the thermodynamic data. This verifies the accuracy and reliability of this new method.

Reference

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