Reactivity of ceramic coating materials with uranium and uranium trichloride

Sung Ho Lee*, Choon Ho Cho, Yoon Sang Lee, Han Soo Lee, Jeong-Guk Kim
Korea Atomic Energy Research Institute, P.O. Box, 105, Yusong, Taejon 305-600, korea
(shlee6@kaeri.re.kr)

1. Introduction

Uranium and uranium alloys are typically induction melted in graphite crucibles under a vacuum. The graphite crucible is used for the manufacturing of uranium ingots in the casting equipment. But, due to the chemical reactivity of uranium and most alloying elements with carbon, a protective ceramic coating is generally applied to the crucibles [1-2]. In this study, to investigate the most suitable ceramic coating material applied to graphite melting crucibles and ingot molds-used in the melting and casting of uranium in the casting equipment, firstly, the thermodynamic analysis was performed by using HSC software to investigate the reactivity between uranium and several ceramic materials and the experiments on the reaction of ceramic coated crucibles in molten uranium were carried out at 1300°C.

2. Experimental

HSC Chemistry software was designed for various simulation and modeling applications based on independent chemical reactions. Using thermodynamic databases, HSC makes conventional thermodynamic calculations**.**

In order to theoretically investigate the thermodynamic reactivity between uranium and several prospective coating materials, firstly, the values of Gibb's free energy for the reaction of ceramic coated crucibles in molten uranium were calculated by using HSC software designed for various simulation and modeling applications based on independent chemical reactions using thermodynamic databases. The experiments are carried out in a high frequency induction coil furnace under a dynamic vacuum better than 10^{-3} Pa obtained by an diffusion pump. The experimental apparatus is shown in Fig. 1.

3. Result and Discussions

Reactivity of uranium in the ceramic coated crucibles can be thermodynamically evaluated by calculating the values of Gibb's free energy according to the temperature because a spontaneous reaction is occurred at negative values of Gibbs free energy. The values of Gibbs free energy for the reactions of ceramic coated crucible in liquid uranium and uranium trichloride were calculated by using the HSC software. The possible reactions between uranium and ceramic coating materials are as follows:

$$Al_2O_3 + 1.5U = 1.5UO_2 + 2Al \quad (1)$$

$$ZrO_2 + U = UO_2 + Zr \quad (2)$$

$$2MgO + U = UO_2 + 2Mg \quad (3)$$

$$2CaO + U = UO_2 + 2Ca \quad (4)$$

$$Y_2O_3 + 1.5U = 1.5\ UO2 + 2Y \quad (5)$$

$$ZrO_2 + Y_2O_3 + 2.5U = Zr + 2Y + 2.5UO_2 \quad (6)$$

The possible reactions between uranium trichloride and ceramic coating materials are also as follows:

$2CaO + UCl_3 = 3CaCl2 + UO_2 + UO$ (1)

$3MgO + 2\ UCl_3 = 3MgCl_2 + UO_2 + UO$ (2)

$Y_2O_3 + 2\ UCl_3 = 2YCl_3 + UO_2 + UO$ (3)

$ZrO_2 + UCl_3 = ZrCl_3 + UO_2$ (4)

$ZrO_2 + Y_2O_3 + 3\ UCl_3 = ZrCl_3 + 2YCl_3 + 2UO_2 + UO$ (5)

Fig. 2 shows the chemical stability of oxide materials with uranium according to the temperature. As shown in Fig. 2, From the results of the thermodynamic analysis, the reactivity of the other ceramic materials except for alumina and zirconia with uranium may not be found in the temperature range of concern. Fig. 3 shows the chemical stability of oxide materials with uranium trichloride according to the temperature. From the thermodynamic calculation results**,** calcium oxide and magnesium oxide may react with the uranium trichloride, however, the other coating materials do not react with the uranium thrichloride.

4. Conclusions

Based on the results from the thermodynamic analysis by using HSC software and reaction experiments, yttria and yttrium stabilized zirconia (YSZ) are shown as the most suitable coating materials. Yttria provides superior protection above 1300°C, however, YSZ becomes less satisfactory above 1200°C when directly applied on graphite [2].

From these results, yttria was selected as the most suitable ceramic coating material applied to graphite crucibles in the U casting equipment

Fig.1. Experimental apparatus.

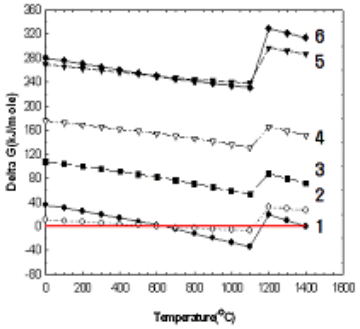

Fig. 2. Chemical stability of oxide material with uranium .

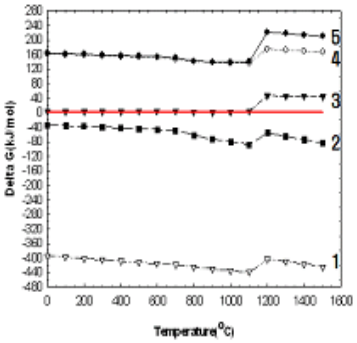

Fig. 3. Chemical stability of oxide materials with uranium trichloride .

REFERENCES

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