Melting temperature measurement of UO_2-Gd_2O_3 fuels

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1. Introduction

UO_2-Gd_2O_3 fuel is widely used as a burnable absorber in light water reactors. The melting temperature of oxide fuel is one of the most important properties for estimating the behavior of nuclear fuel during both normal operation and postulated accident conditions.

Sintering a powder mixture of UO_2 and Gd_2O_3 produces the UO_2-Gd_2O_3 fuel pellets. This fuel is a solid solution that Gd ions replace U ions in the lattice. The melting of a solid solution generally occurs in a range of temperature; it begins at the solidus temperature and ends at the liquidus temperature. Beals et al. [1] reported that the solidus temperature of UO_2-Gd_2O_3 was 300-400℃ lower than the liquidus temperature. However, Wada et al. [2] found out that the solidus temperature was very close to the liquidus temperature although they did not measure the solidus temperature. Thus, there is a big difference between the known solidus temperatures of UO_2-Gd_2O_3.

In this study, the melting temperature of commercial UO_2-Gd_2O_3 fuel pellets imported from the commercial fuel manufacturer were measured by thermal arrest method and compared with the recommended melting point used in fuel design & analysis models.

2. Experiments

Commercial UO_2 fuel pellets containing 4, 6, and 8wt% Gd_2O_3 were broken into fragments. About 10g of each fuel fragments were loaded in a tungsten capsule, and then the capsule was evacuated down to 10^{-2} Torr and filled with helium.

A special long hole is made in the central position of the capsule. This hole can provide the black body condition for measurement of temperature. The capsule was surrounded by carbon fiber. This arrangement was placed in the center of induction coin in a chamber, which was evacuated below 10^{-4}Torr and then purged by Ar gas. The capsule was heated by an induction generator. The temperature of fuel was continuously measured at the black body hole using a pyrometer mounted on the chamber. The temperature obtained by the pyrometer was calibrated against the known melting points of materials such as Nd_2O_3 and Gd_2O_3.

The power for heating the capsule was steadily increased to certain power using computer program, and thereby the fuel temperature was increased in a simplar way. The temperature where a rising temperature was arrested by the heat of fusion was determined as a melting temperature.

3. Results

Table 1 shows the pellet properties used in melting point measurement experiments.

Table 1. Comparison of the pellet properties and manufacture processes of commercial UO_2-Gd_2O_3 fuels.

Manufacturer		KAERI	B	C
Gd_2O_3 contents(wt%)		6wt%	4,6,8wt%	6wt%
Process	Powder	milling	mixing	2step milling*[1]
	Dopant (ppm)	Al (100)	X	Al(100)-Si(40)
	Sintering temperature/ time	1730°C/4hr	1740°C/4~6.5hr	1780°C/6hr
	Sintering atmosphere (H_2O/H_2)*[2]	3%	5%	3%
Pellet density (g/cm^3)		10.33	10.22	10.25

*[1] UO_2 mill and then UO_2-Gd_2O_3 mixture mill.
*[2] $Po_2(H_2O/H_2) \approx Po_2(CO_2/H_2)$

Fig. 1 shows a typical example of temperature-time profile for Gd_2O_3 melting. The arrest of temperature rise was detected near the known melting temperature 2420°C of Gd_2O_3.

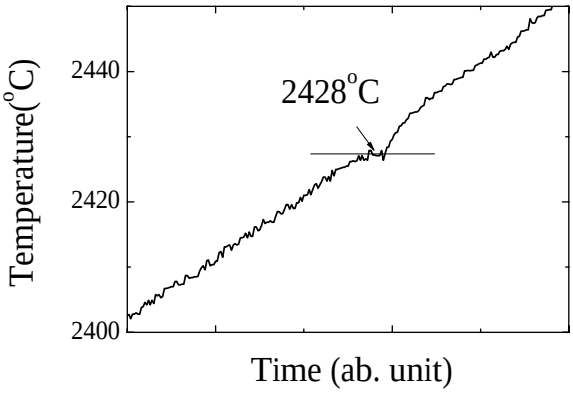

Fig.1. Heating curve for Gd_2O_3 reference sample

Fig. 2 shows an example of temperature-time profiles for the UO_2-8wt%Gd_2O_3 fuel. The temperature profile shows a slow-down in the slope of increasing

temperature in the range between 2500 and 2600 . In addition, the typical thermal arrest is appeared in the vicinity of 2725 . The UO_2-8wt%Gd_2O_3 fuel is a solid solution and thus has both solidus and liquidus temperatures. The solidus temperature (T_S), start of melting, can be identified by a decrease in the temperature slope, and the liquidus temperature (T_L), end of melting, can be identified by an increase in the temperature slope. The arrows in Fig. 2 indicate the determined T_S and T_L. The T_S is difficult to determine since the change in temperature slope is blunt.

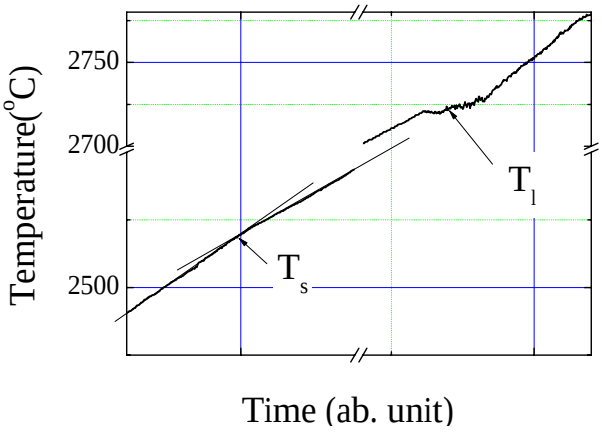

Fig. 2. Heating curve for UO_2-8wt%Gd_2O_3

The determined melting points of commercial UO_2-Gd_2O_3 fuels are plotted in Fig. 3 together with recommended melting points used in the fuel design & analysis models for comparison. The determined T_S and T_L are almost linearly decreased as the Gd_2O_3 content increase, and they are slightly higher than the recommended data. But both decreasing rates of T_S and T_L according to the Gd_2O_3 content are in good agreement with those of recommended lines.

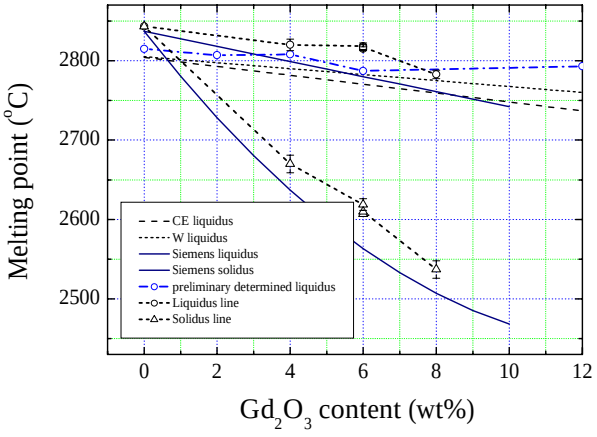

Fig.3. Determined melting temperature of commercial UO_2 -Gd_2O_3 fuels with content variation

Fig. 4 shows the polished microstructure of UO_2-6wt%Gd_2O_3 fuel after the melting experiment. We can find the large columnar grains and cracks, which were caused by thermal gradient and volume contraction at the freezing stage.

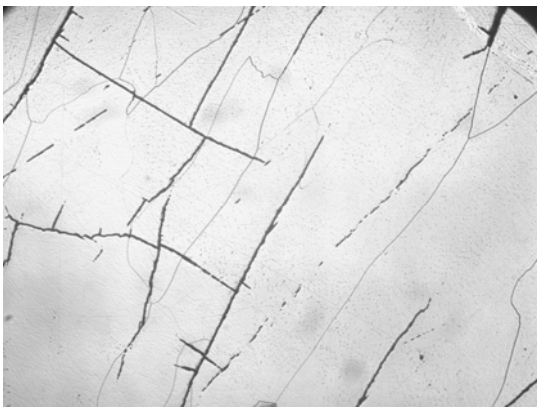

Fig. 4. Microstructure of UO_2-6wt%Gd_2O_3 fuel after the melting experiment.

4. Conclusion

The melting temperatures of commercial UO_2-Gd_2O_3 fuels were measured by thermal arrest method using the high frequency induction-heating tool. The solidus temperature (T_S) and liquidus temperature (T_L) variation according to the Gd_2O_3 content are determined as follows;

$$T_S(^{\circ}C) = 2842(11) - 46(6) \times x \ \ (wt\%\ Gd_2O_3)$$

$$T_L(^{\circ}C) = 2847(9) - 6(1) \times x \ \ (wt\%\ Gd_2O_3)$$

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