The Lattice Parameter Measurements using X-Ray Diffractometry for (U, Ce)O_2 and UO_2+5wt%CeO_2+Nd_2O_3

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1. Introduction

UO_2+5wt%CeO_2 sintered pellets have been used for simulating the Pu-bearing mixed oxide fuel (MOX). In order to evaluate and predict the behavior of MOX fuel in reactor, the thermophysical properties of sintered pellet should be measured and assessed [1-3].

In this respect, the lattice parameter and the theoretical density are fundamental and useful data. In this experiment, the lattice parameter was measured as a function of Ce and/or Nd contents using X-ray diffractometry (XRD) using CeO_2 as a surrogate of PuO_2, and the fitted equations for relationship between the lattice parameter and the composition were obtained. Finally, the theoretical density was calculated by using these results.

2. Methods and Results

2.1 Specimen preparation

UO_2+ x mol%CeO_2 (x=0, 7.63, 14.84, 21.68 and 28.17) and UO_2+ 5 wt%CeO_2+ y mol%Nd_2O_3 (y=0, 1, 3.94, 5 and 10) sintered pellets were prepared as follows.

UO_2 (BNFL, IDR-UO_2), CeO_2 (Aldrich, 99.9%) and Nd_2O_3 (Aldrich, 99.99%) powders were mixed with various Ce and/or Nd contents using a Turbula™ mixer for 1h. Then the powder mixtures were milled using a Dynamic Ball Milling Apparatus (DM) for 4h. Milled powder mixture was compacted with a compaction pressure of 300 MPa and sintered at 2023 K for 4h in a flowing H_2 atmosphere.

2.2 Lattice parameter measurement and theoretical density calculation

X-ray diffraction peak of a sample was measured by XRD (MAC-M03XHF) using a Cu-Kα target. The step scanning method was used (counting time=5 sec., step width=0.05°). Samples for the X-ray diffraction were cut to axial direction from a sintered pellet and polished. To avoid any possibility of a slight oxidation during crushing, carefully polished samples were used in this experiment.

The XRD pattern of UO_2+5wt%CeO_2+y mol%Nd_2O_3 is shown in Figure 1. It shows that the measured peak is gradually shifted to the right side with increasing Nd contents. But as the CeO_2 and Nd_2O_3 forms a solid solution in the UO_2 matrix, the shape of peak pattern was not changed in the whole range (20~120°). The results of UO_2+x mol%CeO_2 have a similar tendency.

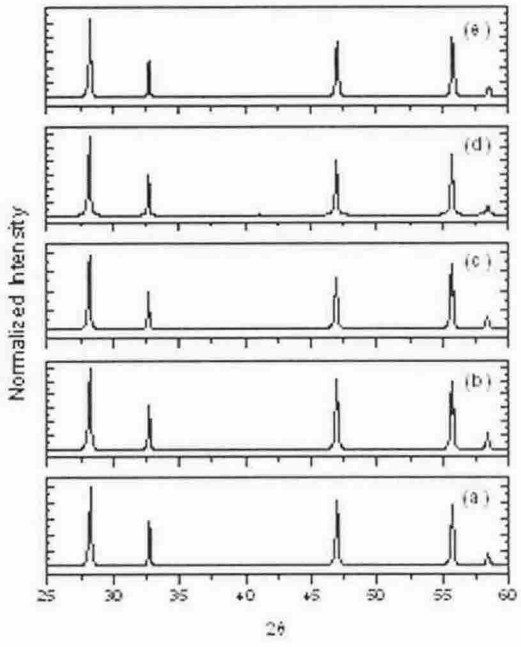

Figure 1. XRD pattern of the (U, Ce)O_2+ y mol%Nd_2O_3 samples with the Nd content: (a) y=0, (b) 1, (c) 3.94, (d) 5 and (e) 10.

Figure 2 shows the results of a calculated lattice parameter using the measured XRD peak pattern of UO_2+x mol%CeO_2 and UO_2+5wt%CeO_2+y mol%Nd_2O_3. The lattice parameter was calculated by using a Nelson-Riley method from XPRESS™ program (X-ray powder research software).

The measured lattice parameter decreased with increasing Ce and/or Nd contents. These results were in good agreement with the reference data [4-6]. And their lattice parameter linearly decreased and followed the Vegard's law. That is to say, in the range of these results, CeO_2 and Nd_2O_3 were completely dissolved in UO_2 matrix. The contraction of UO_2+5wt%CeO_2+Nd_2O_3 is slightly larger than that of (U,Ce)O_2. Additionally, it could be considered that the O/M ratio measured for these samples would be the near-stoichiometric state at these Ce and/or Nd contents

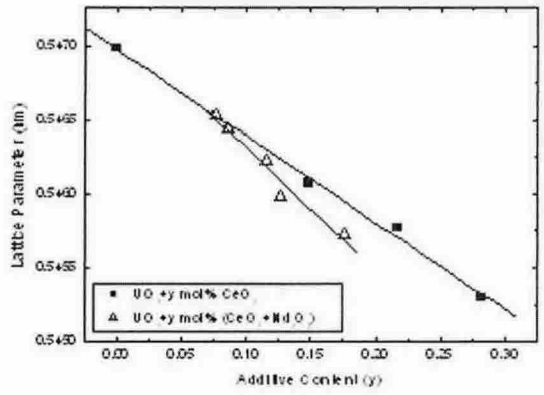

Figure 2. The measured lattice parameters of (U, Ce)O_2 and (U, Ce)O_2+ Nd_2O_3 as a fuction of the Ce and Nd contents.

The following equation for UO_2+CeO_2 was obtained for the relationship between the lattice parameter a in pm and the CeO_2 content, with the [Ce] (mol%) in the range from 0 to 28.17:

$$a = 0.54695 - 0.00582 \times [Ce] \quad (1)$$

Similar equation for UO_2+5wt%CeO_2+Nd_2O_3 was fitted using the same method in the range from 0 to 10:

$$a = 0.54651 - 0.00829 \times [Nd] \quad (2)$$

Finally, the theoretical density was calculated by using eq. (1) and (2). In this calculation, it is assumed that UO_2+CeO_2 and UO_2+5wt%CeO_2+Nd_2O_3 have a cubic structure, base on the similarity of XRD peak pattern.

Table 1. The calculated theoretical density from the measured lattice parameter using eq. (1) and (2).

Sample	Ce or Nd content (x, y, mol%)	Theoretical density (g/cm^3, 100%TD)
UO_2 +x mol%CeO_2	0	10.96005
	7.63	10.68293
	14.84	10.41944
	21.68	10.16860
	28.17	9.92952
UO_2+5wt%CeO_2 +y mol%Nd_2O_3	1	10.61489
	3.94	10.41525
	5	10.34314
	10	10.00202

Table 1 shows that the calculated theoretical density decreased with increasing Ce or Nd contents. Although the contraction of lattice parameter, the mass difference between the dopant and U affect the reduction of theoretical density.

3. Conclusion

The lattice parameters of (U, Ce)O_2 and UO_2+5 wt%CeO_2+Nd_2O_3 pellet specimen were measured by using X-ray diffraction, and the theoretical density was calculated from the measured lattice parameter. As results, the lattice parameter of both samples linearly decreased, and their theoretical densities were reduced due to the mass difference as well.

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