Burnup Determination of Dry Process Refabrication Fuel by Using Neodymium Isotope Monitors

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1. Introduction

One of the important parameters required for studies of irradiated nuclear fuel is burnup, which is the number of fission per 100 heavy nuclide atoms initially present in the fuel. Destructive method, which is based on the determination of specific nuclides by chemical analysis after appropriate separation procedures, is widely used as a reference method to measure the burnup of irradiated fuel[1]. The isotope $^{148}$Nd was selected mainly because its fission yield is independent of the fissioning actinide, and because of its low thermal neutron capture cross section. An approach is to use another monitor such as the sum of $^{145}$Nd and $^{146}$Nd because it appears invariant with neutron flux and fluence.

In order to check the consistency of post-irradiation analysis results, correlations between parameters of irradiated nuclear fuels such as concentration of heavy elements and fission products, ratios of their isotopes and burnup were established[2,3]. These correlations can be used to identify reactor fuels and to estimate the burnup and Pu production. Some of these correlations may also be useful for safeguards purposes.

The aim of this work is to determine the total burnup by using various Nd monitors on the same sample from SIMFUEL and dry process refabrication fuels irradiated in Hanaro reactor and to compare the results for the validity of the methods. In addition, the dependences of U, Pu and Nd isotope composition on burnup values, and correlations between U and Pu and their isotopes for these fuel samples were characterized experimentally.

2. Methods and Results

2.1 Irradiated Fuel Dissolution and Sample Preparation

A irradiated fuel sample weighed exactly was placed in a dissolution flask of dissolution apparatus. The fuel sample was refluxed for more than 10 hours in HNO$_3$(1+1) without catalyst. This fuel solution was weighed and an aliquot was diluted with the HNO$_3$(1+1) with the aid of ORIGEN calculation for the estimation of nuclides content in the irradiated nuclear fuel. An aliquot of the diluted fuel solution was placed in a capped vial and transferred from the shielded facility into a glove box.

2.2 Separation Procedure

Chemical separation was carried out for both the unspiked and the spiked($^{233}$U, $^{242}$Pu and $^{150}$Nd) sample solutions in the same experimental conditions in glove box without heavy shieldings. Two portions were subjected to determine U, Pu and Nd isotopes in sample with and without spike addition followed by two sequential anion exchange separation procedures shown in reference [3,4].

2.3 Determination of Isotopic Composition

The U, Pu and Nd fractions collected from the spiked and unspiked fuel samples were prepared for mass spectrometric determination. In this work, all measured average ratios of Nd were corrected for mass discrimination to achieve high accuracy on burnup measurement. After the mass spectrometric measurement and the correction for their isotope compositions of U, Pu and Nd portions isolated from a spiked and a unspiked sample solution, the concentrations of U, Pu and Nd in sample solution were determined by the isotope dilution method.

Table 1. Total Burnup Determined by Nd-148 Method for the Irradiated SIMFUEL and Dry Process Refabrication Fuel Samples

<table>
<thead>
<tr>
<th>Fuel</th>
<th>MWD/MTHM</th>
<th>Irradiation</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Nd-148 (a)</td>
<td>Calc. (b)</td>
</tr>
<tr>
<td>D4C1</td>
<td>41,798±1,306</td>
<td>42,200</td>
</tr>
<tr>
<td>D4C2</td>
<td>3,308±103</td>
<td>3,300</td>
</tr>
<tr>
<td>D4C3</td>
<td>31,154±974</td>
<td>30,600</td>
</tr>
<tr>
<td>D3C</td>
<td>39,869±1,246</td>
<td>38,900</td>
</tr>
</tbody>
</table>

D4C2 : SIMFUEL
b : Calculated from ORIGEN-2 code

2.4 Determination of Effective Fission Yield and Burnup

In this work, burnup value(in atom % fission) of the irradiated SIMFUEL and dry process refabrication fuel by various Nd isotope monitors was calculated by a
procedure in reference[1]. Neodymium-148, the sum of
$^{145}$Nd and $^{146}$Nd, and the sum of total Nd isotopes($^{143}$Nd,
$^{144}$Nd,$^{145}$Nd, $^{146}$Nd, and $^{150}$Nd) were used as a
fission monitor. The successful application of this
technique requires accurate measurements of the fission
product monitor and heavy atoms and an accurate value
for the effective fission yield. The effective fission yield
was calculated from the weighted fission yields averaged
over the irradiation period by the methods in
reference[4,5]. Table 1 gives the total burnup(atomic %
fission) determined experimentally by Nd-148 method
and calculated from ORIGEN-2 code for each fuel
samples. The data in Table 1 were in agreement within
2.4%. Table 2 gives the total burnup measured by various
monitors, $^{145}$Nd + $^{146}$Nd and $^{148}$Nd isotopes. The data by
two methods were in agreement within 1.3% for the same
fuel sample.

Table 2. Comparison of Total Burnup Determined by
Different Nd isotope Monitors for the Irradiated
SIMFUEL and Dry Process Refabrication Fuel Samples

<table>
<thead>
<tr>
<th>Fuel</th>
<th>Nd-148 (a)</th>
<th>Nd+$^{145+146}$ (b)</th>
<th>Diff. (b/a)</th>
<th>Irradiation</th>
</tr>
</thead>
<tbody>
<tr>
<td>D4C1</td>
<td>41,798</td>
<td>41,656</td>
<td>0.997</td>
<td>Gori-1+Hanaro</td>
</tr>
<tr>
<td>D4C2</td>
<td>3,308</td>
<td>3,264</td>
<td>0.987</td>
<td>Hanaro</td>
</tr>
<tr>
<td>D4C3</td>
<td>31,154</td>
<td>30,808</td>
<td>0.989</td>
<td>Gori-1+Hanaro</td>
</tr>
<tr>
<td>D3C</td>
<td>39,869</td>
<td>39,695</td>
<td>0.996</td>
<td>Gori-1+Hanaro</td>
</tr>
</tbody>
</table>

D4C2 : SIMFUEL

2.5 Correlation between Burnup and Isotope
Compositions

In this work, It was evaluated that the dependences of
various isotope ratios for U, Pu and Nd against burnup,
and correlations between isotope themselves, e.g.
$^{234}$Pu/$^{238}$Pu and total burnup, $^{235}$U/$^{238}$U and total burnup,
$^{236}$U/$^{238}$U and total burnup, $^{235}$U/$^{238}$U and $^{236}$U/$^{238}$U,
atom % U isotope and total burnup(Figure 1), atom % Pu
isotope and total burnup(Figure 2), $^{148}$Nd/$^{145}$Nd and total
burnup, $^{144}$Nd/$^{145}$Nd and total burnup, $^{142}$Nd/$^{145}$Nd and total
burnup, atom % $^{238}$U and $^{144}$Nd/$^{145}$Nd, atom % $^{239}$Pu and $^{143}$Nd/$^{145}$Nd. The isotope ratios determined
experimentally were expressed with good linearity against
the total burnup and isotope compositions from
experimental data.

3. Conclusion

The use of all the Nd isotopes in the determination of
burnup for nuclear fuel has the advantage of confirming
the value obtained for $^{148}$Nd. No additional separation

work or mass spectrometric analysis is needed. The
agreement of the number of fissions calculated from
isotopes sensitive to fuel composition, confirms the fissile
isotope content. The Nd isotope pattern provides
information on the real irradiation characteristics which is
necessary for evaluating the fuel performance in the
reactor.

Figure 1. The Dependence of U Isotopes on Total Burnup(Ft).

Figure 2. The Dependence of Pu Isotopes on Total Burnup(Ft).

REFERENCES