Neutron Diffraction Analysis of Mechanically Milled Spent Fuel

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1. Introduction

The DUPIC(Direct Use of spent PWR fuel in CANDU reactor) nuclear fuel is an advanced nuclear fuel cycle to recycle fissile material in the pressurized water reactor(PWR) spent fuel for use in the heavy water reactor(CANDU). Simulated spent fuel is used to investigate the fabrication process and properties of the DUPIC fuel using natural uranium oxide and stable isotopes of fission product elements. The simulated spent fuel contains about 3 wt% of fission product elements such as Zr, Mo, Nd, Ru and Ce. Mechanical milling is used to fabricate fine and homogeneous distribution of the fission product element in the simulated DUPIC fuel. The crystallite size of powder is important because it affects fabrication properties such as sintering behavior. In this study, the size of milled powders of the simulated spent fuel was measured from the line broadening of neutron diffraction profiles.

2. Experimental procedures

The mixed UO_2 and fission product oxides powders were dry-milled in an attritor with a rotation speed of 200rpm, ball-to-powder ratio of 4:1 for 30, 60, and 120 minutes. The simulated DUPIC fuel powders were compacted into pellets under a pressure of 3 ton/cm^2 and 4 pellets were stacked in a vanadium can for neutron diffractometry by the high resolution powder diffractometer of the HANARO research reactor in the Korea Atomic Energy Research Institute. The wavelength of neutron beam was 0.18348 nm, and the monochrometer was a Ge(331) single crystal. The diffraction patterns were obtained from 20 to 155° by 2è interval of 0.05°, and the maximum intensity was about 20000 counts.

3. Results & Summary

Diffraction line broadening due to crystallite size was measured using various techniques such as the Stokes' deconvolution, profile fitting methods using Cauchy function, Gaussian function, and Voigt function, and the Warren-Averbach method. Voigt function showed the best fitting for the neutron diffraction peak of mechanically milled spent fuel. The non-uniform strain, stacking fault and twin probability were measured using the information from the diffraction pattern. The realistic crystallite size could be obtained after separation of the contribution from the non-uniform strain, stacking fault and twin. The average crystallite size was slightly decreased from 61.7 nm to 39.6 nm with increasing milling time from 30 min to 120 min according to profile fitting using Voigt function followed by Warren-Averbach methods. The crystallite size distributions of mechanically milled spent fuel became narrower as milling time increased. Line broadening analysis of mechanically milled spent fuel is effective in estimating crystallite size distribution statistically.