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Standardless Quantitative Analysis of PWR Spent Fuel with WDS-SEM

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1. Introduction

Electron beam microanalysis has been applied to study the behavior of many fission products (FPs) and transuranium elements inside a nuclear spent fuel (SF) [1]. Recently, a Wavelength Dispersive Spectroscopy (WDS) has been installed on the shielded Secondary Electron Microscopy (SEM) at the PIE facility of KAERI. The preliminary tests on the quantitative analysis of various SF elements have been carried out.

Two main points for the WDS analysis of spent fuels should be considered. The first is the background increase due to the gamma ray emitted from samples. The second is that a standard material of a few elements is difficult to be obtained. For instance, plutonium standard is very sensitive material according to the nuclear non-proliferation, and xenon is hard to form a compound as noble and inert gas.

This paper proposed the standardless peak centering and quantification methods of WDS.

2. Methods and Results

In this section some of the techniques which are need to standardless quantification analysis are described.

2.1 Shielded SEM

WDS analyses were performed with a NORAN IbeX (WDS) and a shielded Philips XL-30 (SEM) as in figure 1. These systems are shielded by 17 cm Carbon steel and confined by hardened glass to protect against a radiation exposure to an operator.

Fig. 1. Shielded SEM.

2.2 Standardless peak centering

The characteristic X-ray energy measured in WDS is determined by the geometric positions of the motor-driven detector and diffractor of WDS on the Rowland circle. Such positions corresponding to X-ray energy should be offset. There are two methods on peak calibration of standardless elements as follows;

- Interpolation among offset values of the adjacent characteristic X-ray energy peaks to a target X-ray line
- Adoption of offset of the similar energy peak to a target

X-ray line interference is frequently occurred among the M-lines of the actinides. The Pu Mα1 line interferes with the U Mβ line [2]. Hence the Pu Mβ line is suitable when the PWR spent fuel will be analyzed by WDS. Cd Lβ2 (3.528 keV, relative intensity: 15) line was adopted in order to calibrate Pu Mβ (3.531 keV) line. In case of xenon, Sb Lβ2,15 (4.101 keV, r.i.: 17) was adopted for Xe Lα1 (4.110) line.

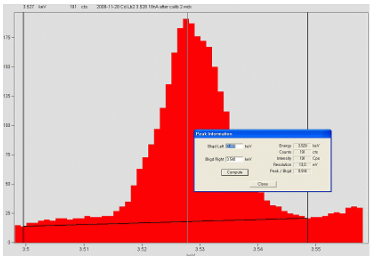

Fig. 2. Peak centering of Cd Lβ2 line.

2.3 Shielding of WDS

The X-ray detector of the WDS is a gas proportional counter, which has low detection efficiency for a high energy photon like gamma ray [3]. But FPs and actinides contained at lower 1 wt% in the SF sample has a feature of a very low peak to background ratio. Therefore a straight path between a sample and a detector must be shielded to make the P/B ratio high.

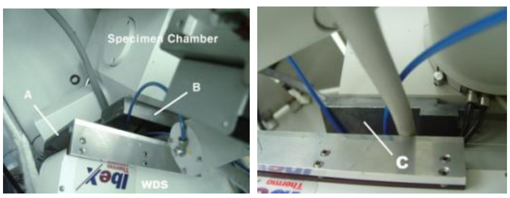

Fig. 3. Shielding between WDS and chamber.

Table I: Shielding effect for the background

Element	*X-ray line*	*Energy, keV*	*Crystal*	*Bragg angle*	*Counts per second*		
					Non-shield	*C type*	*B+C type*
Nd	$L\alpha1$	*5.229*	*LiF*	*36.07*	*1173*	*224*	*114*
U	$M\alpha1$	*3.171*	*PET*	*26.57*	*751*	*143*	*87*
Pu	$M\beta1$	*3.531*	*PET*	*25.06*	*694*	*181*	*110*
Xe	$L\alpha1$	*4.109*	*PET*	*23.68*	*1001*	*239*	*133*

Figure 3 shows lead shielding type for WDS. Through the combination of B and C type, background counts due to gamma-ray minimized by 100 counts as table I.

2.4 Beam and specimen current

The electron beam stability is important for the quantitative analysis. It directly affects the measurement precision. To inspect the beam stability, the picoammeter (Keithley 6485 model) was installed. The beam current was measured in a Faraday cup and then the target was changed to the PWR spent fuel. Sum of backscattering and secondary electron coefficient was 0.46 under the following conditions;

- Electron gun: LaB6
- Accelerating voltage: 25 kV
- Filament current: 1.82 A
- Spot size: 8.2

2.4 Standardless quantification

Standard intensity can be calculated by the Monte Carlo simulation code. But, it is difficult to evaluate diffraction and detection efficiencies of WDS because of the complexity. Therefore, standard intensity needs to be measured at the adjacent X-ray line to the target X-ray line. If the simulation coupled with the measurement, those efficiencies can be evaluated.

3. Conclusions

Standardless quantification analysis using WDS-SEM is the useful technique to measure the standardless elements inside PWR spent fuel. In the future, we will focus on evaluating the standard intensity of xenon and plutonium, and trying to measure the secondary election yield by applying about +50V bias voltage to the sample stage.

REFERENCES

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