Characteristics of X-ray fluorescence of nuclear materials

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1. Introduction

THE low energy X-ray measurement system is used for on-site analytical measurements of safeguards in various nuclear facilities, such as nuclear power plant and reprocessing plants. Specially, the hybrid system of X-ray absorption spectrometry (XAS), such as L-edge densitometry (LED) [1, 2], and X-ray fluorescence spectrometry (XRF) has an important role of safeguards for nuclear facilities. LED is a technique of determination of uranium concentration as a continuous X-ray energy beams transmit a uranium liquid sample for safeguards. Compared to K-edge densitometer [4], due to relatively lower energy (L-edge energy is 17.17 keV) of Uranium L series energy than K-series energy, L-edge densitometer does not require high purity germanium detector with liquid nitride cooling. Therefore, the L-edge densitometer is appropriate for portable equipment for on-site nuclear material inspection and safeguards at facility sites. XRF combined with LED is a technique of finding of nuclear materials from reflected characteristic X-ray photons. In this study, characteristics of XRF of nuclear materials are simulated Monte Carlo method (Geant4 [3]) for feasibility of the system for determination of concentration of nuclear species. The analysis method of uranium concentration or minor actinides is applied using combination of linear extrapolation from jump of L-edge of sample and ratio between uranium and minor actinide from XRF measurement.

2. Methods and Results

2.1 LED/XRF system

The integrated LED/XRF system consists of an X-ray tube, shields, a sample container, and detectors. The schematic figure is shown as Fig. 1. The sample container, quartz cuvette, has 2 mm optical path length and 17.5 × 3.5 mm^2 area. The detectors are set up front of sample container for L-edge densitometry and X-ray tube direction with an angle for XRF. Both of detectors are assumed pure silicon where 28 mm^2 × 500 μm for simplification of the simulation because the doping concentrations are low and do not affect the deposition energy calculation. The X-ray spectrum incident to sample solution is used an experimentally measured spectrum data to avoid difficulties of X-ray generation simulation. The diameter of X-ray photon starting point to the sample is 1.6 mm in front of collimator with hole of 1.6 mm-diameter and length of 7-cm-long. The X-ray cone angle is 1 ° from extraction of the off-axis photons by the collimator. The X-ray voltage and current are from 30 kV/100 μA and silver anode X-ray generator [6]. The spectrum range is 0 ~ 30 keV because of measurement of uranium L series < 22 keV including K_α and K_β line of silver anode, and continuous energy components by bremsstrahlung.

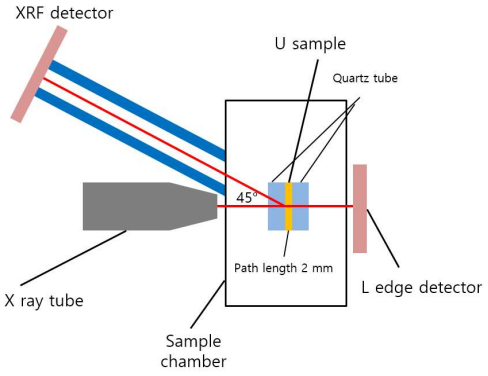

Fig. 1 Schematic figure of integrated L-edge densitometer with X-ray fluorescence system (LED/XRF)

2.2 Concentration from X-ray fluorescence spectrometry

In order to determine concentrations of minor actinide, such as, plutonium and americium, in samples of fission products or reprocessing, the analysis requires X-ray fluorescence spectrometry (XRF) techniques. For feasibility of the simulation, Monte Carlo simulation is performed for 0.2g/cm^3 liquid uranium at 10 ~ 60° of the angle opposite to the sample cuvette. For simulation feasibility, the ratio of intensity of the first order characteristic X-ray line energies, $L_{\beta 2}/L_{\alpha 1}$, is 0.215 corresponding to reference value 0.209 from intensities for uranium in Table I.

TABLE I. CHARACTERISTIC X-RAY LINE ENERGIES OF NUCLEAR MATERIALS*

Element (Level)	Energy (keV)	Intensity (eV/h)
U ($L_{\alpha 1}$)	13.61	2.411
U ($L_{\beta 2}$)	16.43	0.506
Pu ($L_{\alpha 1}$)	14.28	2.652
Pu ($L_{\beta 2}$)	17.26	0.564

*Experimental data from Handbook of X-ray Data by G. Zschornack, Springer, 2007 [5]

XRF spectrometry is useful to analyze minor actinides, for example plutonium in mixture of nuclear materials rather than only a uranium sample. The simulated XRF spectrum is shown in Fig. 2. 14.28 keV energy peak which corresponds to $L_{\alpha 1}$ characteristic X-ray line of plutonium is observed.

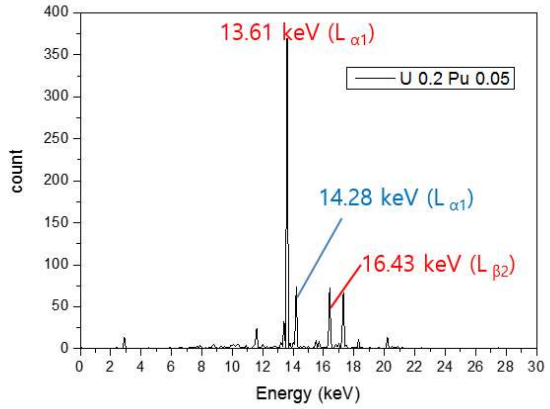

Fig. 2 Simulated XRF spectrum of LWR solution (U 0.2 g/cm^3 in 3M HNO_3 solution) with small amount of plutonium (0.05 g/cm^3). 14.28 keV peak of $L_{\beta 2}$ of plutonium is shown in the spectrum. (XRF detector angle is 45°.)

The amount of plutonium can be calculated by following equation.

$$\frac{M_{Pu}}{M_U} = \frac{C_{Pu}}{C_U} \times \frac{E_U}{E_{Pu}} \times \frac{I_U}{I_{Pu}} \times \frac{A_{Pu}}{A_U} \quad (1)$$

Uranium concentration M_U is obtained from L-edge densitometry analysis. I_U and I_{Pu} are intensities of uranium and plutonium in Table II. C is count number of detector of and E of each element is efficiency. The amount is also calculated using relative atomic mass (A in the equation). The estimated plutonium from the XRF spectrum is shown in Table II.

TABLE II. ESTIMATED PLUTONIUM CONCENTRATION FROM SIMULATION*

Input (g/cm^3)	Estimation (g/cm^3)
0.01	0.0093
0.05	0.041
0.10	0.082

* Uranium concentration is 0.2 g/cm^3.

The estimated Pu concentration from the simulation has the difference of ~ 20 % from input concentration except for 0.01 g/cm^3. The discrepancy is caused by volume effect of emission of characteristic X-ray line of nuclear materials. The XRF photon from the surface after reaction with incident X-ray photon emits without any obstacles. The volume effect can be uncertainty for determination of nuclear materials The XRF system is necessary to improve in order to minimize volume effect.

2.3 Effect of the angle of the detector

In order to maximize counting efficiency, the determination of angle of the detector is required since the XRF peaks have low counting efficiency compared to L-edge spectra. As shown in Fig. 3, total counting efficiency maximizes at low angle due to the square geometry of the sample container. However, the angle is determined at 20 ~ 30°. The X-ray tube and collimator geometry has to be considered. The estimated Pu concentration is not change for the angle as shown in Fig. 4. The angle does not affect characteristics of XRF except for counting efficiency.

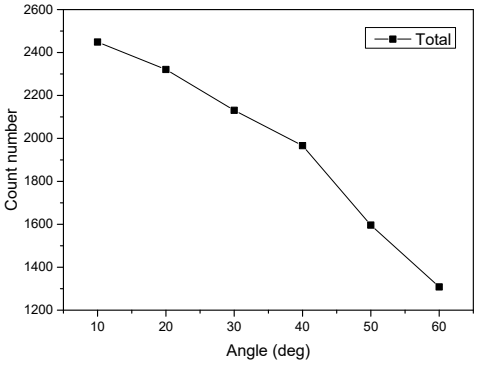

Fig. 3 Total counting efficiency of XRF peaks for the detector angle

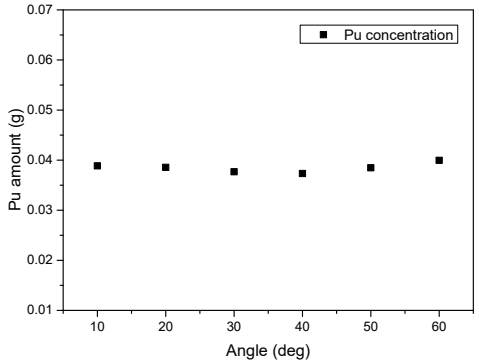

Fig. 4 Pu concentration from XRF analysis for the detector angle

3. Conclusions

In this study, The XRF characteristics was simulated from Monte Carlo method. The peaks were obtained from nuclear material mixture. The estimated nuclear material concentration is low due to the volume effect of the sample. The correction factor or minimization of the effect is required. The geometrical effect of counting efficiency was investigated. The lower angle detector, the higher counting efficiency could be obtained, but the estimated concentration did not change at any angles. Based on the simulation and design, the LED/XRF system will be fabricated in the further study.

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