

Nuclear Fuel Assay through analysis of Uranium L-shell by Hybrid L-edge/XRF Densitometer using a Surrogate Material

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Abstract—Assay of L-series of nuclear material solution is useful for determination of amount of nuclear materials and ratio of minor actinide in the materials. The hybrid system of energy dispersive X-ray absorption edge spectrometry, i.e. L-edge densitometry, and X-ray fluorescence spectrometry is one of the analysis methods. The hybrid L-edge/XRF densitometer can be a promising candidate for a portable and compact equipment due to advantage of using low energy X-ray beams without heavy shielding systems and liquid nitrogen cooling compared to hybrid K-edge/XRF densitometer.

A prototype of the equipment was evaluated for feasibility of the nuclear material assay using a surrogate material (lead) to avoid radiation effects from nuclear materials. The uncertainty of L-edge and XRF characteristics of the sample material and volume effects was discussed in the article.

Index Terms—L-edge, XRF, Nuclear Material Solution, X-ray absorption, X-ray fluorescence

I. INTRODUCTION

THE low energy X-ray measurement system is intended to be used for on-site analytical measurements of safeguards in various nuclear facilities, such as nuclear fuel fabrication facilities and reprocessing plants. Specially, the hybrid system of X-ray absorption spectrometry, 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. Compared to K-edge densitometer [3], due to relatively lower energy (L_{III}-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 nitrogen cooling and heavy shielding system for shielding of K-series radiation (~ 115 keV) of uranium. Therefore, the L-edge densitometer is appropriate for a portable equipment for on-site nuclear material inspection and safeguards at facility sites. XRF

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combined with LED is a technique of characterizing the element content of nuclear materials from fluoresced characteristic X-ray photons.

In this study, the hybrid L-edge/XRF densitometer (HLED) was developed for determination of nuclear materials concentration. We verified that the system has feasibility of determination of the concentration of nuclear material from analysis of a surrogate material such as lead nitrate (Pb(NO₃)₂) solution prior to the work of actinide-bearing materials.

II. THE HYBRID LED/L-XRF DENSITOMETER

The schematic figure of HLED is shown in Fig. 1. The equipment was fabricated based on previous study [4] of Monte Carlo simulation for the instrument design and feasibility study of determination of nuclear material concentration. The prototype of hybrid L-edge/XRF densitometer (HLED) consists of an X-ray tube, shields, a sample container, and two detectors. Both of detectors are silicon drift detectors (SDD) of 25mm² × 500 μm which is XR-123SDD by Amptek™ including a digital pulse processor. The X-ray tube of 1.6 mm anode spot diameter is fabricated in Moxtek™ with maximum power, voltage, and current of 4 W, 50 kV, and 200 μA in front of collimator of a hole diameter of 0.5 mm and length of 5-cm-long. The sample container is made of Teflon to protect corrosion by nitric acid with 2 mm-optical path length and 300 μl volume. The windows material is Mylar® (polyester) film of 6 μm to maximize detection efficiency. There are beam lines for detection of transmission and XRF. The XRF detection beam line is aligned with 45 degrees for the sample window normal and the transmission detection line set up in the direction of the X-ray beam as shown in Fig. 1. The X-ray tube is operated by 30 kV and 90 μA with rhodium (Rh) anode.

X-ray photons from the tube transmit or randomly react to the sample by XRF process. The transmission and fluoresced photon energy spectra are recorded by the detectors. The rhodium anode with characteristic peaks of K_α (20.21 keV) and K_β (22.72 keV) can be a role of calibration of energy below 30

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keV as well as generation of continuous X-ray spectrum, i.e. bremsstrahlung.

The experiments were performed for lead nitrate ($\text{Pb}(\text{NO}_3)_2$) solution with 0.01, 0.05, 0.1, and 0.2 g/cm^3 to avoid radiation effects for the performance evaluation of the equipment. The blanket material is water in order to obtain transmission.

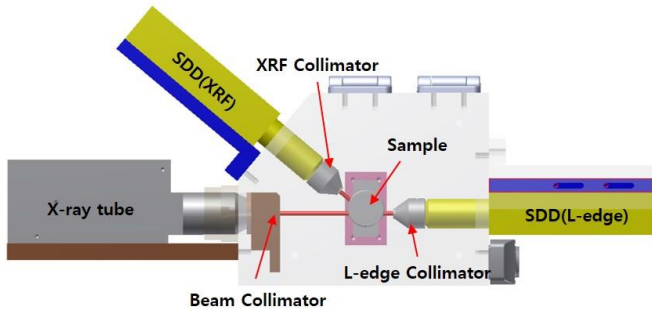


Fig. 1 Design of Hybrid L-edge/XRF Densitometer

III. EVALUATION OF HLED

A. L_{III} -edge Analysis

The discontinuous curves, i.e. jumps, of transmission spectrum are observed. The height of the jump of the spectrum determines the sample concentrations. Material concentrations through transmission spectrum can be calculated by the following equation (1).

$$\rho_{\text{Pb}} = \frac{\ln[T(E_-)/T(E_+)]}{\Delta\mu\Delta} \quad (1)$$

$T(E_-)$, $T(E_+)$ are transmission at the energies E_- (lower than L_{III} -edge) and E_+ (upper than L_{III} -edge). D is sample thickness which is exactly called optical path length and $\Delta\mu$ is mass attenuation coefficient difference at upper and lower L_{III} -edge. Extrapolated fitting in linearized a representation of $\ln(I/T)$ - $\ln(E)$ is applied for determining upper and lower transmission for L-edge. Fitting intervals from 11 – 12 keV for E_- and 14 – 15 keV for E_+ .

Relative standard deviation (RSD) represents uncertainty of the measurement as shown in Fig. 2. The samples are measured for 3×1000 s respectively. The RSD reduces at high concentration ($> 0.01 \text{ g}/\text{cm}^3$) and slightly increases at $0.2 \text{ g}/\text{cm}^3$. The total counts at high concentration are relatively low compared to low concentration. The result implies that low transmission above $0.2 \text{ g}/\text{cm}^3$ results in low accuracy. As a result, the optimized concentration can be considered above $0.01 \text{ g}/\text{cm}^3$ and below $0.2 \text{ g}/\text{cm}^3$ of lead. The possible concentration range have consistency with the previous study for L-edge of uranium and plutonium [5].

B. X-ray fluorescence of surrogate material

In order to determine concentrations of minor actinide or impurities in a nuclear material sample, the analysis requires

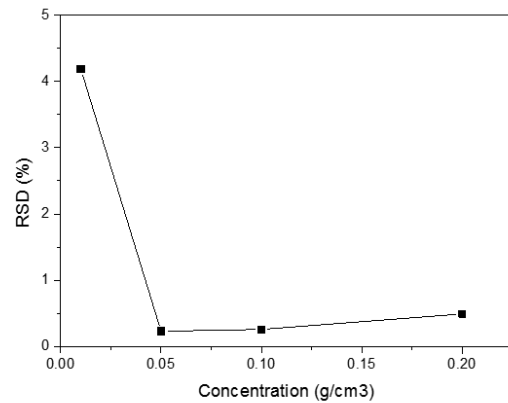


Fig. 2. Relative Standard Deviation (RSD) for each concentration

not only L-edge densitometry but also X-ray fluorescence spectrometry.

The typical XRF peaks are L_α (10.4 keV) and L_β (12.6 keV) of lead. The ratio of L_α and L_β for any concentration is shown in Fig. 3. XRF photons created in the sample are absorbed and interact with lead. The ratio decreases as the concentration increases because the lower energy photons can be more affected by the sample. The relation of the peak intensity and the peak ratio between the concentrations of a sample have to be considered of the XRF analysis for mixed solution samples. Unlike a single element analysis, the analysis for mixed solution samples are required calibration data for target materials using various ratio of elements and concentrations.

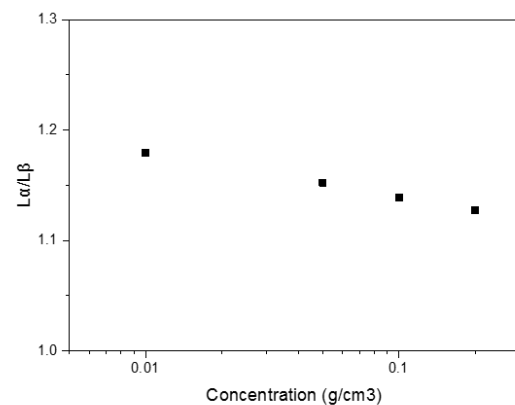


Fig. 3 The peak ratio of L_α and L_β of lead for each concentration

IV. CONCLUSION

The prototype of a hybrid L-edge/XRF densitometer (HLED) was developed for nuclear material solution assay. The equipment has feasibility due to the uncertainty from L-edge analysis near ITV (International Target Value). The evaluation verified that the concentration and volume effects must be considered for assay of multi-element solution samples.

In future work, based on the study, nuclear material will be analyzed and uncertainty will be reduced by analysis of non-radioactive and radioactive standard samples such as lead nitrate and uranium nitric acid solutions.

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