Uranium Solution Assay by L-edge Densitometer

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1. Introduction

Hybrid system of X-ray absorption spectroscopy such as L-edge densitometry (LED) [1, 2] and X-ray fluorescence spectrometry (XRF) is an useful technique of assay of uranium concentration for safeguards. Compared to K-edge densitometer [3], due to lower L-edge energy of 17.17 keV of uranium than K-edge energy of 115.6 keV, L-edge densitometer does not need a high purity germanium detector with liquid nitrogen cooling and heavy shielding systems. Therefore, the L-edge densitometer can be appropriate for portable equipment for on-site nuclear material inspection and safeguards. XRF with LED is a technique of simultaneous measurement of the ratio of nuclear materials in mixture samples from emission of characteristic X-ray photons.

In this study, a portable hybrid L-edge/XRF densitometer (HLED) was developed for determination of concentration of nuclear materials. We evaluated the performance of the equipment as measuring uranyl nitric acid solution with various concentrations.

2. Results and Discussion

2.1 Hybrid L-edge Densitometer

The hybrid LED/XRF densitometer (HLED) consists of an X-ray generator, shields, a sample cell, and detectors. The detectors are set up at front and normal of a sample cell for L-edge densitometry. Both of detectors are silicon drift detectors (SDD) which 25 mm^2 × 500 μm. The diameter of X-ray photon starting point to the sample is 1.6 mm in front of collimator with hole of 0.5 mm-diameter and length of 5-cm-long. The sample cell consists of a 45 ml volume for XRF to maximize detection efficiency with a small volume of 1.5 mm optical path length. There are two beam lines from the X-ray generator to each sample volume as shown in Fig. 1 The X-ray generator is usually operated by 35 kV/100 μA with rhodium (Rh) anode.

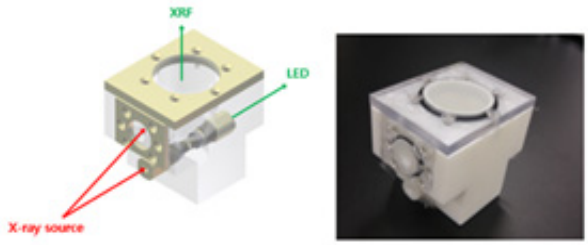

Fig. 1. The beam lines across a sample cell of HLED.

2.2 Results

The fully oxidized U_3O_8 nitric acid solution samples with concentration of 0.01, 0.05, 0.1, and 0.2 g/cm^3 were prepared for experiments. The discontinuous spectrum at near 17.17 keV L_{III}-edge are observed compared to reference spectrum of nitric acid solution in Fig. 2 The height of the jump of the spectrum determines the uranium concentrations.

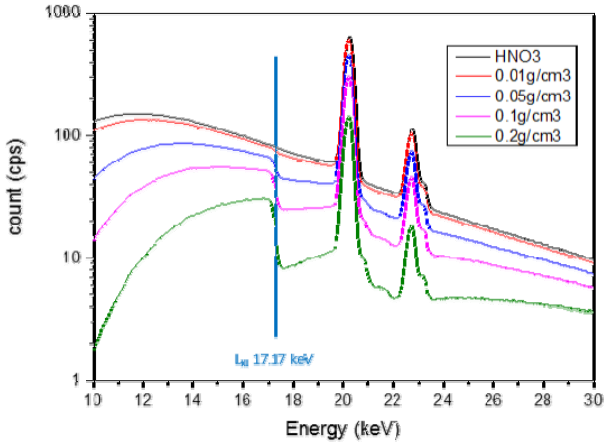

Fig. 2. Transmission spectrum across U_3O_8 nitric acid solution.

The concentration can be obtained by following equation (1).

$$\rho = \frac{\ln(T_- / T_+)}{\triangle \mu D} \quad (1)$$

T_- and T_+ are transmission at the energies E_- (lower than L_{III}) and E_+ (upper than L_{III}) respectively. D is sample thickness (exactly, optical path length) and $\Delta\mu$ is mass attenuation coefficient difference at L_{III}-edge. The transmission of L_{III}-edge is determined by extrapolation fitting in linearization of the upper side and the lower side transmission of L-edge in lnln(1/T)-ln(E) curve. Fitting intervals range from 15 - 16 keV in the lower side and 18 - 19 keV in the upper side of L-edge

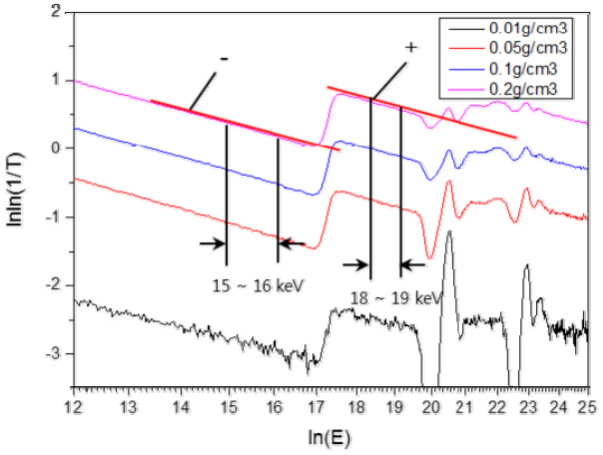

Fig. 3. Analysis of extrapolation fitting in lnln(1/T)-ln(E).

The uranium concentration in U_3O_8 nitric acid solution is shown in Fig. 4 The ratio of uranium in U_3O_8 is usually 83.8%. The analyzed concentrations of uranium are near 84% except for 0.01 g/cm^3. The lnln(1/T) curve of 0.01 g/cm^3 has noise near L-edge as shown in Fig. 3 The background radiation and the short optical path length can affect the transmission of low concentration. The uranium concentration has linearity for concentrations of U_3O_8 with R^2 > 99%.

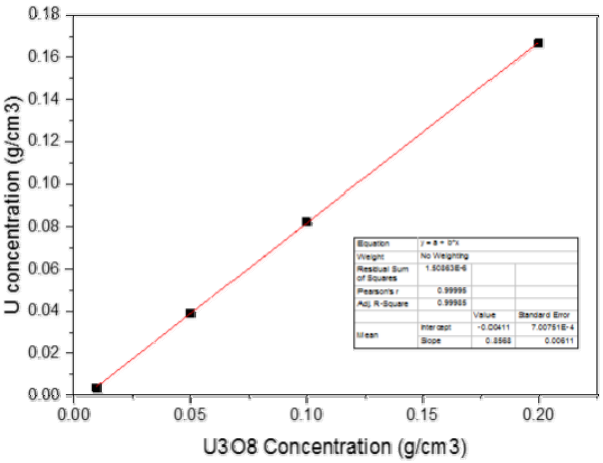

Fig. 4. The uranium concentration in U_3O_8 nitric acid solution.

3. Conclusion

From this study, the proto-type of a hybrid L-edge/XRF densitometer (HLED) was developed and evaluate performance for nuclear material solution assay. Based on the study, the uncertainty of the measurement will be analyzed by analysis of non-radioactive and radioactive standard samples of lead and uranyl nitric acid solutions. The performance of XRF and simultaneous measurement of XRF and L-edge will be evaluated by mixture solution of nuclear material and impurities.

4. REFERENCES

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