

## Multi-element solution L-XRF analysis using the hybrid L-edge/L-XRF densitometer

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### 1. Introduction

The hybrid L-edge/L-XRF densitometer (HLED) was developed for on-site nuclear fuel assays intended for safeguards purpose. The HLED can simultaneously measure both X-ray photon transmissions and characteristic X-ray emissions, which characterizes the elemental composition of samples of interest to determine the concentration of actinide-bearing materials, such as plutonium and uranium, in a nuclear fuel.

From the previous study [1], a prototype of HLED equipment was fabricated and verified by determining the concentration of a surrogate material, a lead nitrate ( $\text{Pb}(\text{NO}_3)_2$ ) aqueous solution.

In this study, in order to mimic mixture of nuclear materials, we have tested the mixture of surrogate material solution of lead nitrate ( $\text{Pb}(\text{NO}_3)_2$ ) and gold(III) chloride trihydrate ( $\text{HAuCl}_3 \cdot 3\text{H}_2\text{O}$ ) aqueous solution for the XRF analysis of multi-element solution.

### 2. System Description

Figure 1 shows the HLED design [1]. The equipment is fabricated based on results from a previous study [2], in which the instrument's design was determined using Monte Carlo simulation. The prototype of HLED consists of an X-ray tube, shielding, a sample container, three collimators and two silicon drift detectors (SDD). The HLED sample container is made of PTFE to protect it from corrosion by nitric acid. The sample container has an optical path length of 2 mm and a volume of 50  $\mu\text{l}$ . The sample container's window material is a 6- $\mu\text{m}$ -thick Mylar® (polyester) film used to maximize detection efficiency. In the system, there are beam lines to detect transmission and XRF emission. The XRF detection beam line has a 45° alignment to the sample window's normal, and the transmission detection line is parallel to the X-ray beam as shown in Figure 1. The X-ray tube is operated at 30 kV and 90  $\mu\text{A}$  with a rhodium (Rh) anode.

X-ray photons from the X-ray tube either transmit through the specimen or interact with it via following three processes for the range of photon energies below 100 keV, elastic scattering, inelastic scattering, or absorption. The transmitted and fluoresced photon energy spectra are recorded by the detectors.

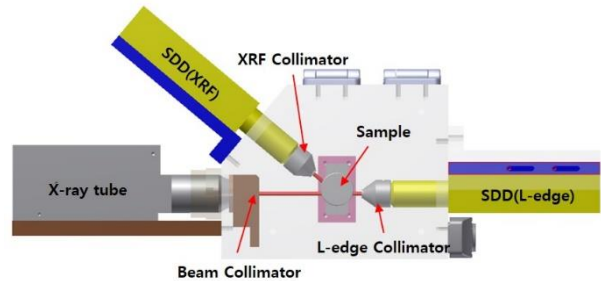


Fig. 1. Design of hybrid L-edge/L-XRF densitometer [1]

### 3. Experimental methods and Results

Experiments were performed for the mixed solution of lead nitrate ( $\text{Pb}(\text{NO}_3)_2$ ) and gold(III) chloride trihydrate ( $\text{HAuCl}_3 \cdot 3\text{H}_2\text{O}$ ) solution with a constant concentration of 0.2  $\text{g}/\text{cm}^3$  of lead nitrate and concentrations of 0.01, 0.02, 0.04, and 0.1  $\text{g}/\text{cm}^3$  of gold(III) chloride trihydrate. The surrogate materials are chosen to only analyze L-edge and XRF characteristics without radiation effect by nuclear materials. Water is a blanket material to obtain transmission. Each sample was measured three times for 1000 s.

Determining minor element concentrations or concentrations of impurities in mixture samples, requires both L-edge analysis and XRF spectrometry. The typical XRF spectra of lead  $L_\alpha$  (10.4 keV),  $L_\beta$  (12.6 keV) and gold  $L_\alpha$  (9.7 keV),  $L_\beta$  (11.1 keV) were measured from various gold concentrations. The peaks are shown in Figure 2.

The XRF photons created in the sample are absorbed by lead and interacts with lead. Assuming that the X-ray beam and the detector are on the same side of specimen in a straight line, the original radiation intensity is  $I_{in}$ . The fluoresced intensity  $I_x$  interacts in the volume  $dx$  at a depth of  $x$  in the sample, and it is given by Equation (1). The  $I_{out}$  from the sample surface is the XRF intensity at the detector [3].

$$I_x dx = I_{in} K \exp(-\mu^{in} x) \cdot \exp(\mu^{out} x) dx. (1)$$

Figure 3 shows the relationship between the peak intensities of  $L_\alpha$  and the concentrations of gold. The peak intensity of lead decreases when gold concentration increases while the concentration of lead is constant because the matrix and volume effect of the mixed solutions.

Figure 4 shows the peak ratio between lead  $L_{\alpha}$  and gold  $L_{\alpha}$  for each concentration. This ratio increases as concentrations of gold increases linearly. The relation between the peak intensity with the peak ratio and the concentrations of a target material have to be considered of the XRF analysis for mixed solution samples. Unlike single element analyses, analyzing samples of mixed solutions requires calibration data for the target materials that uses various elemental ratios and concentrations because of the matrix effect. The relation shows that amount of minor element can be deduced knowing major element concentration.

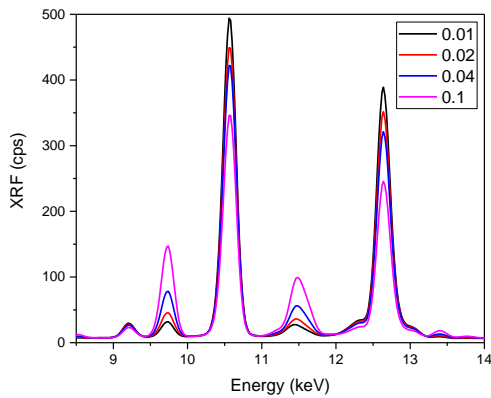


Fig. 2.  $L_{\alpha}$  and  $L_{\beta}$  XRF peaks of a constant concentration of lead and gold in various concentrations of gold

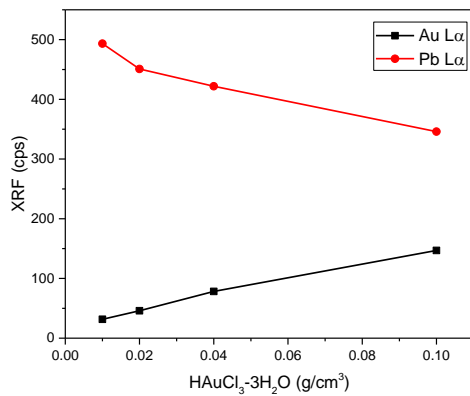


Fig. 3. Peak intensities of gold and lead  $L_{\alpha}$  in various concentrations of gold

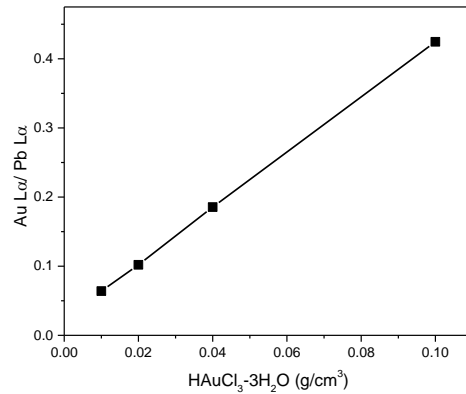


Fig. 4. Gold and lead  $L_{\alpha}$  peak ratios in various concentrations of gold

#### 4. Conclusions

In this study, we investigated the characteristics of XRF peaks for multi-element aqueous solution of surrogate materials prior to nuclear material mixture study. Peak ratio between  $L_{\alpha}$  of gold and lead has linear relation of gold concentration. From this relation, if we know the concentration of lead, the concentration of gold could be calculated by measuring the XRF peak intensity. The effects of concentration of major element and volume must be more considered for more realistic solutions that is, various concentrations of multi-element solution samples.

#### REFERENCES

- [1] S. Joung, and S. Park, Development of a prototype hybrid L-edge/L-XRF densitometer for nuclear fuel assay, Applied Radiation and Isotopes, Vol. 133, p. 81-84, 2018
- [2] S. Park, and J.-K. Shin, S.-W. Kwak, "Development of Hybrid L-edge/XRF Densitometer for Determination of Nuclear Material Concentration", 31 October to 7 November, 2015 IEEE Nuclear Science Symposium & Medical Imaging Conference, 2015.
- [3] D. Reily, N. Ensslin, H. Smih, Jr., and S. Kreiner, Materials. (United States Nuclear Regulat Passive Nondestructive Assay of Nuclear Commission, 1991), 325-327.