Screening Uranium Swipe Samples Using the MMXRF

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1. Introduction

With the purpose of environmental sampling for safeguards (ESS), the Korea Institute of Nuclear Non-proliferation and Control (KINAC) has been established Korean environmental sampling programme in collaboration with the Korea Atomic Energy Research Institute (KAERI) and the Institute for Transuranium Elements (ITU) [1].

For ESS, samples are collected from nuclear facilities and then screened in various measurement equipments such as X-ray fluorescence spectrometry (XRFS), high resolution gamma-spectrometry (HRGS) and alpha/beta spectrometry as well as detailed measurements by bulk and particle analysis using thermal ionization mass spectrometry (TIMS) and scanning electron microscopy (SEM) [2-4].

In this study, we developed the monochromatic micro-focusing X-ray fluorescence (MMXRF) which is focused on uranium detection. The MMXRF can show better results in uranium detection than the existing X-ray fluorescence (XRF). MMXRF has the improved sensitivity since it can reduce background in X-ray scattering under the fluorescence peaks by using micron-sized monochromatic X-ray beam which is generated by doubly curved crystal optics (DCC) [5].

In this study, the results will be described using real uranium swipe samples.

2. Experimental Methods and Results

2.1 Detection Process of MMXRF System

The MMXRF consists of a X-ray tube, a DCC, a XYZ stage, a silicon drift detector (SDD) for detecting secondary X-ray, and a multi channel analyzer (MCA). Characteristics of the MMXRF are shown in table I.

Table I: The characteristics of MMXRF

Nominal Output Power	25 W
Target Material	Ag
Detector Type	SDD
Energy Range	2 ~ 35 keV
Active Detector Area	20 mm^2
Window Material	Beryllium
P/B Ratio	Up to 6000/1

Figure 1 shows the detection process of uranium swipe sample using MMXRF.

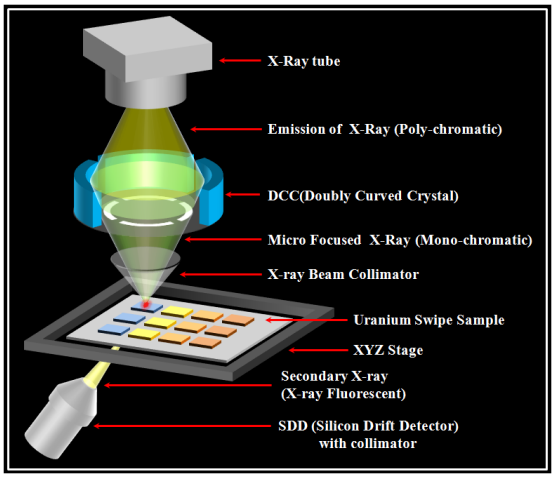

Fig. 1. Detection process of uranium swipe sample using MMXRF system.

2.2 Calibration of MMXRF System

To estimate a uranium mass of an unknown swipe sample, the measurement system has to be calibrated using standard material with same match. 6 reference uranium swipe samples made up of SRM 3164 (Standard Uranium Solution, NIST) and cotton swipes (TX304, ITW company) was fabricated with a size of 10 mm x 10 mm. Each sample contains different mass of uranium: 10, 20, 30, 40, 50, and 70 ng per cm^2.

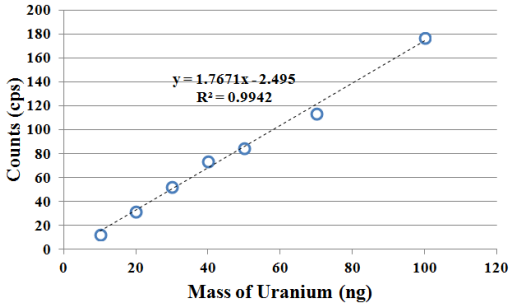

Fig. 2. Calibration curve of the MMXRF with irradiation range 3 mm x 3 mm.

Figure 2 shows the linear relationship between the uranium mass and counts obtained from those reference samples on the MMXRF system. The detection range of the MMXRF was set up 3 mm x 3 mm and this value was applied to all samples. As a result, correlation coefficients (R^2 value) were calculated in 99.4% and the linear function derived from the experience results is shown in equation 1.

$$y = 1.7671x - 2.495 \quad (1)$$

Based on the linear function, consequently, we can estimate uranium mass of unknown environmental swipe samples.

2.3 Sampling and Measurement Methods

We collected uranium swipe samples from 8 points at KAERI using 10 cm x 10 cm cotton swipe as shown in Figure 3.

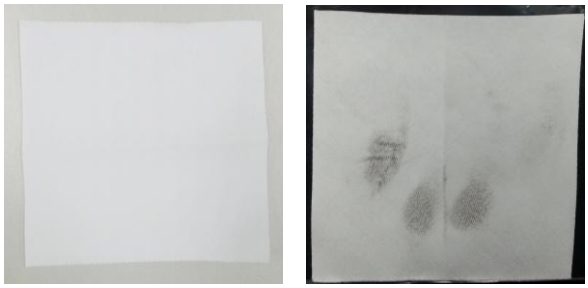

Fig. 3. A new (left) and swiped (right) cotton swipe used for environmental sampling at KAERI

The collected uranium swipe samples were analyzed by the MMXRF to estimate the uranium mass on them. The whole experimental setups for these unknown samples were same that of calibration: detection range was 3 mm x 3 mm and fast screening (point detection) time was 5 sec. Considering the size of the cotton swipe (10 cm x 10 cm), the total measurement time per sample was about 2 hr.

2.4 Analysis of Environmental Uranium Swipe Samples

Figure 4 shows the screening result obtained from one of the collected uranium swipe samples. The results of 2D mapping with counts were identified with the real swipe.

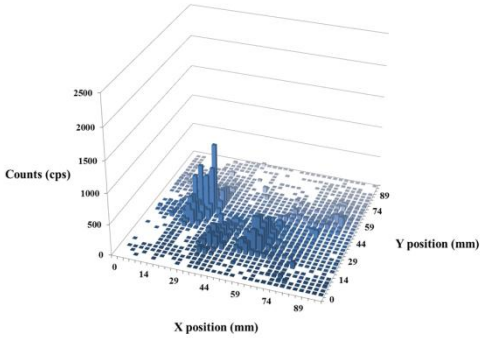

Fig. 4. Screening result obtained from one of uranium swipe samples

Table II shows screening results of all uranium swipe samples collected from 8 points at KAERI. Each swipe sample was analyzed by the MMXRF and the uranium mass was finally calculated by Equation 1.

For in-depth analysis, we selected samples showing the highest values in each point and sent to the ITU for the particle analysis.

Table II. Total screening results of environmental uranium swipe samples collected in KAERI.

(Unit : ng)

Sample no. \ Place no.	1	2	3	4
1	593.6	628.7	588.5	701.8
2	644.9	653.1	605.9	696.0
3	639.2	608.3	1673.3	623.6
4	653.5	601.9	638.0	1068.3
5	649.3	689.5	633.7	766.7
6	629.5	660.2	661.3	910.6
Sample no. \ Place no.	5	6	7	8
1	733.5	21372.9	686.6	2395.2
2	695.4	43879.7	643.3	1893.5
3	836.2	13063.7	670.0	900.3
4	739.3	47296.7	630.3	803.4
5	1075.7	11721.3	660.9	1269.0
6	832.4	19574.5	723.5	1032.5

3. Conclusions

In this study, we draw the calibration curve to estimate a uranium mass of an unknown swipe sample. As a result, calibration curve well matched with calibration curve equation.

Based on the calibration results, the real uranium swipe samples collected in KAERI was analyzed using the MMXRF. In conclusion, we could estimate uranium mass of the real uranium samples. And we select samples showing the highest values in each point to analyze in detail. Selected samples will be sent the ITU for the particle analysis.

In further study, those screening results will be compared with ITU analysis results to evaluate the MMXRF in detail.

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