

## SCREENING AND RADIOMETRIC MEASUREMENT OF ENVIRONMENTAL SWIPE SAMPLES

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### Abstract

Environmental swipe samples are taken by IAEA Safeguards inspectors inside nuclear installations such as enrichment facilities and facilities with hot cells. These samples are screened by radiometric methods before further processing in order to establish the identity and quantity of radioactive isotopes present. High resolution gamma spectrometry is used to measure the concentrations of fission and activation products in radioactive samples as a measure of the burn-up of the fuel handled in the inspected facility. X-ray fluorescence spectrometry is applied to screen for uranium presence in low activity samples. Subsampling of swipes taken inside hot cells is also performed using adhesive carbon discs followed by alpha/beta counting. The sample preparation, measurement and data evaluation procedures are described and examples given of typical results.

### 1. INTRODUCTION

Environmental swipe samples have an activity ranging from 0.001Bq to  $10^7$ Bq. They are deposited either on cotton carriers (10cm X 10 cm) or “hot cell” swipes and may contain a wide range of actinide elements and fission products[1,2]. These samples must be screened by radiometric methods at the Safeguards Analytical Laboratory and the Clean Laboratory before further processing in order to establish the identity and quantity of radioactive isotopes present. The screening procedures are designed to rapidly process large numbers of samples to yield semiquantitative estimates of radioactivity levels and main radiation contributors with attention paid to the isotopes and elements of special interest for Safeguards. Screening measurements are nondestructive to avoid any contamination or cross-contamination of the measured samples. Wide variations in the activity deposition on the sample carrier, isotopes present in the sample and a restricted measurement time do not allow to reach high accuracy in the screening results. Typically measured intensities of the analyzed elements, isotopes etc. are treated with calibrations performed with reference samples which are not identical to the screened items.

### 2. SAMPLE PREPARATION FOR SCREENING

Cotton swipes are packed in double plastic bags. One series number consists of 4 - 5 swipes plus 1 control swipe put in one large plastic bag together. The given swipe series is screened by HRGS either in the conventional nuclear analysis laboratory (NL) or in the Clean Laboratory (CL) depending on the measured total swipe dose rate. All swipes of the series are screened by HRGS together without opening the protective bag. For the measurement in the NL HRGS system swipes are packed in a plastic beaker in a form of a cylinder 6 cm high and 6 cm in diameter. The beaker is placed on the top of the detector head. High active swipes are measured at larger distance between the detector head and the beaker. In the CL HRGS system swipes are fixed in a Marinelli beaker horizontally and the beaker is placed by the sample changer on the top of the detector.

Hot cell swipes are small cellulose wipes in plastic bottles. The collected material is distributed in a circular spot with a diameter of 2.5 cm. Hot cell swipes are screened by HRGS singly without opening the protective bottle. Swipes are placed horizontally on the top of the detector (NL system) or at the center of the Marinelli beaker (CL system).

The XRF screening is applied to detect U, Pu or other elements of interest on the surface of the swipe sample. The most active swipe of the given series is selected, packed in the plastic holder and placed by the sample changer on the top of the XRF analyzer with  $^{109}\text{Cd}$  excitation source.

The alpha/beta screening of the hot cell swipes is performed with specially prepared adhesive sticky carbon discs. Inside a plastic glove box the hot cell swipe is removed from the container and the active surface of the swipe is touched by the sticky surface of the carbon disc. Following this, the active surface of the disc is covered by a 5 micron protective polypropylene film. The opposite sticky side of the disc is attached to a stainless steel planchet. This type of subsampling allows to make measurements with systems where conductivity of the sample is necessary.

### 3. SCREENING MEASUREMENTS

#### 3.1. Gamma screening

Both NL and CL laboratories are equipped with high sensitivity and high performance HRGS spectrometers. The NL gamma spectrometer includes a 42% efficiency HPGe detector, ultra low activity Pb passive shield and active NaI(Tl) guard detector, providing suppression of Compton background in the measured spectrum [3]. The CL gamma spectrometer has a 90 % efficiency detector, ultra low activity Pb shield and sample changer with 15 positions. Both spectrometers were manufactured by CANBERRA Packard. Genie-2000 software provides full operational service beginning from measurement functions, spectra storage, evaluation, calibration functions, isotopes identification and activity evaluation with reporting of the results, including errors and detection limits for the preselected isotopes. Calibration of the spectrometers is performed with reference sources in the energy range from 50 keV to 2650 keV. Measurement time in routine screening is 1 hour. Table I lists reported results of the gamma screening of the sample 46-03. The sample contains 4 swipes with total count rate 18.3 cps measured at a distance of 25 cm between the detector head and the sample. The reported data also includes the confidence level estimated by the software [4]. The last 6 isotopes in the table were not detected, but preselected for the reporting. For these isotopes the minimum detectable activity is given in the last column of Table I.

More than 50 different isotopes were measured in swipe samples by gamma screening. The data obtained are used for estimation of the parameters of the spent fuel handled at the facility. For example, with data given in the Table I it was estimated that at the hot cell facility a high enriched fuel with cooling time about 1 year and irradiation time of about 2 years was handled [5].

Table I. Reported activities for swipe 46-03

Isotope	Conf. level	Activity, Bq/Swipe	Uncertainty, Bq/swipe
Nb-95	0.994	78.7	2.97
Zr-95	0.999	45.7	1.86
Ru-103	0.913	2.98	.645
Ru-106	0.604	120	11.3
Cs-134	0.885	24.5	.972
Cs-137	0.996	1.22	4.46
Ce-144	0.946	1050	34.2
Sb-125			14.2
Ba-140			1.84
Ce-141			5.37
Eu-154			6.79
Eu-155			8.12
Am-241			6.48

### 3.2. XRF screening

The XRF screening device is equipped with a 100 mm<sup>2</sup> Si(Li) detector, annular 20 mCi Cd-109 excitation source and sample changer with 12 positions. An inspected area is about 12 cm<sup>2</sup> at the center of the analysed swipe. The swipe is packed in double plastic bags. Measurement time is 1 hour. The detection limit for U is about 4 micrograms. Elemental composition of the swipe is measured on special analytical request with a PHILIPS 1480 XRF analyser and UNIQUANT software[6]. High absorption of the XRF quanta in the plastic bags does not allow to estimate light elements presence in the swipe, but all the elements with  $Z > 10$  can be detected. Typical elements detected are lead, iron, silica, titanium. A new XRF system is under development now, which will allow to make XRF scanning of the whole swipe surface. The system includes 2 semiconductor detectors, a 100 W X-Ray tube and a robot arm.

### 3.3. Alpha/Beta screening

Three devices can be used for alpha/beta screening of the adhesive sticky carbon discs prepared by subsampling of hot cell swipes (see section 2.):

- I. Grid Ionisation Chamber IN-614, INTERTECHNIQUE [7]
- II. TENNELEC Alpha/Beta Low level counter LB-5500, Oxford Instruments Inc [8]
- III. Alpha spectrometer 7401 with PD-100-13-100AM detector, CANBERRA Packard [9]

The measured characteristics of the devices are listed at the Table II. The efficiencies were

Table II. Efficiency and background data

Device	alpha efficiency, %	beta efficiency, %	alpha backgr, cps	beta backgr, cps
IN-614	41	11	0.37	1.1
LB-5500	27	50	0.01	0.13
Alpha PIPS	3.6	0.01	0.00003	0.0011

measured with reference alpha and beta point sources covered by a 5 micron polypropylene foil. The energy range for alpha counts was taken from 3.0 MeV to the maximum energy where counts were observed (~ 8 MeV). The energy range for beta counts was taken from 0 to 2 MeV. For the ALPHA PIPS system the minimum energy could not be measured below 550 keV because of electronics restrictions.

Twelve routine hot cell swipe samples were measured to estimate performance of the screening systems. The measurement time of the prepared sticky carbon discs was 1 h for IN-614 and LB-5500. For the ALPHA PIPS system, measurement time was up to 24 h because of the low efficiency. Tables III and IV list results of measurements. The last row of the Tables III and IV lists the correlation coefficients between activities measured with different systems. The data presented demonstrate that:

- All three systems can be used for identification of beta signatures.
- Quantitative beta activity results can be obtained from LB-5500 and IN-614 measurements.
- ALPHA PIPS system can only indicate the presence of beta emitters.
- LB-5500 has poor performance for the detection of alpha emitters. The reason is that real swipes contain strong beta emitters and subtraction of the overlapping beta pulses from the alpha channel using cross talk function [8] makes a large error in the alpha particle count rate.
- IN-614 can be considered as an appropriate candidate for the routine screening both alpha and beta emitters.

- ALPHA PIPS can be used on special analytical request to detect low alpha activity (less than 0.01 alpha/sec).

Table III. The measured beta activities (beta/sec).

Sample	LB 5500	IN-614	Alpha PIPS	LB/IN ratio
78-03	81591	107515	1060585	0.76
85-01-01	0.15	0.36	0.00	0.43
85-02-01	8203	8185	769	1.00
89-01	6.71	4.69	0.00	1.43
89-01-02	0.44	0.52	0.00	0.84
90-01	345	453	16.94	0.76
90-02	3201	3124	1144	1.02
91-01	357	276	54	1.29
91-02	1303	765	123	1.7
96-01	19.6	25.7	1.5	0.76
96-02	1.2	0.69	3.3	1.8
96-03	11030	15474	7259	0.71
Correlation coefficient	LB $\diamond$ IN 0.9996	IN $\diamond$ Alpha 0.989	LB $\diamond$ Alpha 0.988	Average ratio 1.04+/-0.42

Table IV. The measured alpha activities (alpha/sec).

Sample	LB 5500	IN-614	Alpha PIPS	Alpha/IE ratio
78-03	17523	3602	2916	
85-01-01	-0.01	0.05	0.0005	
85-02-01	53	0.03	0.0233	0.697
89-01	-0.19	0.02	0	
89-01-02	-0.04	0.05	0	
90-01	20.6	0.02	0.00067	
90-02	64	0.39	0.27834	0.707
91-01	-3.3	0.07	0.05633	0.813
91-02	-51	0.07	0.05195	0.765
96-01	0.66	0.05	0	
96-02	-0.07	0.02	0.00175	
96-03	888	0.06	0.00117	
Correlation coefficient *)	LB $\diamond$ IN -0.001	IN $\diamond$ Alpha 0.981	LB $\diamond$ Alpha -0.088	Average ratio 0.75+/-0.05

\*) Correlation coefficient was evaluated without use of the 78-03 sample data.

#### 4. CONCLUSIONS

A system of analytical devices has been applied for the screening of the environmental swipe samples for safeguards. The IAEA Safeguards Analytical Laboratory performs non-invasive radiometric measurements of environmental swipe samples. The results reported are used to specify further detailed analytical treatment of the samples at SAL or network laboratories. New techniques are under development and will be applied as they become available.

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