Determination of Uranium enrichment with FRAM – comparison of electrically cooled Germanium detector Detective200 and U-Pu detector

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Abstract: In the context of fissile material detection, the determination of Uranium enrichment and of the Plutonium isotopic vector is tremendously important. The negligence of such information may lead to drastic if not fatal consequences to the general public in case the material in question is used in explosive devices such as an Improvised Nuclear Devices (IND). The implemented automatic analysis routine of the electrically cooled Detective 200 detector has been investigated in earlier measurements measuring Uranium and plutonium sources of different enrichment respectively isotopic composition. The results have been partly disappointing. The object of the present work was to investigate the general ability and the quality of the spectra obtained with the Detective200 in comparison to the commonly used U-Pu detectors cooled with liquid nitrogen. To determine the uranium enrichment measurements were carried out with a source of depleted Uranium and different shielding materials. The evaluation of the spectra was performed with the isotopic analysis code FRAM. The results for the Uranium enrichment for the Detective200 system were compared to the enrichment result of the commonly used U-Pu detectors. The results of these measurements and an assessment of the ability of the FRAM software combined with the Detective200 system in determining the Uranium enrichment will be presented.

Keywords: gamma measurement, Uranium enrichment, U-Pu, FRAM

1 Introduction

The detection and identification of radioactive and especially nuclear material are crucial with respect to countermeasures against nuclear terrorism. Uranium and Plutonium play an especially important role in this context as these materials are the core substance of explosive devices such as an Improvised Nuclear Device (IND) which may be used to cause severe or even fatal injuries to the public in comparatively large areas, rendering them uninhabitable as a consequence. To prevent such acts from happening, this material must be tracked either before such devices are assembled of at least before they are ignited.

In order to achieve an identification of said material, highly sensitive gamma detectors with electrically cooled germanium semiconductors are a reasonable choice, primarily because of their excellent gamma energy resolution. U-Pu detectors cooled with liquid nitrogen (LN) are also a good option, although these devices have limitations with regard to on-site measurements due to the size and weight of their cooler and the availability of LN in the field.

The electrically cooled detectors are commonly equipped with an implemented identification routine. The ability of such analysis routines to reliably determine the isotopic composition of Pu or the degree of enrichment of U has been the subject of previous measurements [1, 2]. The results were, at least in part, quite disappointing although the quality of the gamma spectrum would have enabled an

appropriate identification. Therefore the special code called FRAM by Ametek/ORTEC was used for subsequent analysis of the measured spectra regarding uranium enrichment for a comparison of several electrically cooled germanium detectors with another germanium detector cooled with LN.

2 Measurement set-up

A series of measurements with low and high enriched as well as natural and depleted uranium was performed at the Institute for Transuranium Elements (ITU) in Karlsruhe, Germany, as low or high enriched uranium is not available at our institute. Additional measurements were performed with depleted uranium and a sample of mineral containing natural uranium at Fraunhofer INT.

2.1 Measurements at ITU

At ITU, four different uranium samples with different degrees of enrichment were available: 0.3% (depleted), 0.71 % (natural), 4.5 % (low enriched), and 91.4 % (high enriched). As shielding material 2 mm of lead and a combination of 1.5 cm of steel and 5 cm of an explosive simulate were used. The latter contained the same chemical components as explosive without actually being an explosive. This combination of steel and explosive simulate represented the configuration of an IND.

Four different measurement systems with electrically cooled germanium crystals were tested during this series: the Detective EX, the Micro Detective, the Detective 200 (all designed by Ametek/ORTEC), and the FALCON 5000 by Canberra. Figure 1 shows these detectors in a typical setup positioned around a source.

Figure 1: Measurement array with four detectors positioned around an unshielded source; the detectors are (clockwise starting on the bottom left): Detective EX, Micro-Detective, Detective 200 and Falcon 5000.

The main characteristic values of the detectors are given in table 1 as well as the outer dimensions of the systems. The three Detectives (Detective EX, Micro-Detective and Detective 200) differ from each other in respect of the Germanium crystal and the firmware. Detective EX and Micro-Detective are comparable concerning the crystal (see table 1) but differ in the firmware. Detective 200 and Micro-Detective both have the newer firmware V3, but the Detective 200 has a larger crystal.

Detector, Manufacturer	Weight of device [kg]	Size of Device [cm^3]	Crystal Size [cm] ø/Length	Energy Resolution [keV]		Relative Efficiency [%] for ^{60}Co	Battery Life [h]
				at 186 keV	at 662 keV		
Detective EX, ORTEC	12	37 x 18 x 34	5 / 3	1.3	1.7	16	> 3
Micro-Detective ORTEC	7	37 x 15 x 28	5 / 4	1.3	1.7	11	5
Detective 200, ORTEC	21	43 x 24 x 39	8.5 / 3	1.1	1.5	52	3
Falcon 5000, Canberra	15	40 x 35 x 16	6 / 3	1.1	1.4	18.5	6 - 8

Table 1: Overview of the detection systems and their specifications; the weight and the size of the Falcon 5000 do not include a necessary PC; weight and size figures for all systems include batteries; the relative efficiencies are obtained using the standard measurement procedure in which a ^{60}Co source is placed 25 cm away from the end-cap of the detector.

In all cases the spectra can be stored and the spectrum files can be exported and transferred to a specialist. The systems feature automatic identification and in particular special SNM (Special Nuclear Material) modes. The latter have been specially considered for these uranium measurements. In order to gain additional information concerning uranium enrichment and the isotopic composition of plutonium, the data were analyzed with the isotopic analysis software PC/FRAM 5.1 (ORTEC version) [3]. Thereby we used the existing parameter files u_cx_120-1010 for the Detectives and uleu_plnr_060-250 for the U-Pu detector after adapting the energy calibration but without any further optimization.

Compared to the firmware used by the Detective EX, the firmware of the Detective 200 and Micro-Detective has limited information output especially in the standard non-expert mode. To have a better comparability we therefore did not strictly use the recommended settings for the subsequent measurements although we wanted to evaluate the results which non-expert users would gain.

The identification mode displays of the Detective devices continuously update the results and run until they are manually stopped by the user. The Falcon 5000 can be operated with a preselected time or without a preset. However the result of the SNM Mode will be only displayed after the end of the measurement. The chronological development of the measurement results were specially observed for all detectors. Whenever possible, spectra with longer measurement times were obtained for example during midday or night.

2.2 Measurements at INT

These measurements were done to compare the performance of electrically cooled germanium detectors mentioned above to the quality of a U-Pu detector cooled with LN. The U-Pu detector yields gamma spectra in the lower energy region up to 300 keV which is a relevant region for measurements with nuclear material. Such type of detector is part of a wide-range collection of measurement systems at INT for detecting radioactive or nuclear material [4].

Measurements were performed with several detectors: the Detective 200, the Micro Detective, and a U-Pu detector cooled with LN. A 1.8 kg block of depleted uranium and a sample of mineral containing natural uranium were measured, both unshielded and shielded with 5 mm of Fe. Figure 2 shows a typical measurement layout with the depleted uranium shielded by Fe and the Micro Detective.

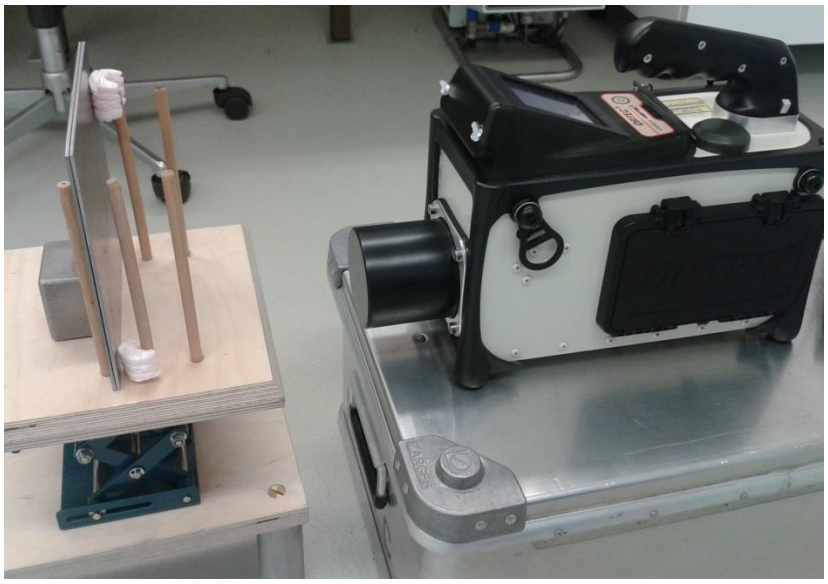

Figure 2: Measurement array with Micro Detective (on the right) and 1.8 kg block of depleted uranium (on the left) shielded by 5 mm of Fe.

3 Measurement Results

3.1 ITU Measurements

With each of the four uranium (depleted: DU, natural: nat. U, low enriched: LEU, and high enriched: HEU) at least one measurement was performed without shielding material and with the combination of steel and explosive simulate. LEU and HEU were additionally examined with lead shielding. Time permitting; multiple measurements were done with the same setup.

Tables 3 to 5 show comparisons of the identification results yielded by the implemented analysis routines of the devices and the results subsequently gained by the analysis with FRAM for the Detective 200, the Detective EX, and the FALCON 5000 for the uranium measurements. The derived results base on the categorization limits in table 2:

Material	Characterization limits
DU	$^{235}U < 0.64$ %
Nat. U	0.64 % < $^{235}U < 0.78$ %
LEU	0.78 % < $^{235}U <$ 20 %
HEU	$^{235}U > 20$ %
RGPu	$^{239}Pu < 90$ %
WGPu	$^{239}Pu > 90$ %

Table 2: Limits for the characterization of uranium and plutonium material

Sample	Shielding	Life Time (s)	Implemented Analysis	Result Derived From FRAM
DU	Unshielded	956	*Nucl. U*	DU
DU	Steel + Simulate	1458	*Nucl. U*	DU
Nat. U	Unshielded	874	*Nucl. U*	*LEU*
Nat. U	Steel + Simulate	1079	*Nucl. U*	*DU*
LEU	Unshielded	1189	*Nucl. U*	LEU
LEU	Steel + Simulate	7765	*Nucl. U*	LEU
LEU	Lead	255	*Nucl. U*	LEU
HEU	Unshielded	1575	*Nucl. U*, HEU	HEU
HEU	Steel + Simulate	691	*Nucl. U*, HEU	HEU
HEU	Lead	57613	*Nucl. U*, HEU	HEU

Table 3: Comparison of Detective 200 analysis results for the uranium measurements yielded by the implemented analysis routine and calculated with FRAM; color code: blue color refers to a correctly identified isotope or material, but the degree of enrichment or categorization is wrong, green color means both the identification and categorization are correct.

Sample	Shielding	Life Time (s)	Implemented Analysis	Result Derived From FRAM
DU	Unshielded	225	DU, elev. U concentration	DU
DU	Steel + Simulate	207	*elev. U conc.*	DU
Nat. U	Unshielded	890	–	nat. U
Nat. U	Steel + Simulate	596	*DU, elev. U conc*	*DU*
Nat. U	Steel + Simulate	712	*DU, elev. U conc.*	*DU*
LEU	Unshielded	1732	LEU, elev. U concentration	LEU
LEU	Steel + Simulate	302	*elev. U conc.*	*DU*
LEU	Steel + Simulate	6425	*nat. U, elev. U conc.*	LEU
LEU	Lead	352	*DU, elev. U conc.*	*DU*
LEU	Lead	152	*DU, elev. U conc*	*DU*
HEU	Unshielded	1004	*LEU*	HEU
HEU	Unshielded	212	*LEU*	*LEU*
HEU	Unshielded	137	*LEU*	*LEU*
HEU	Steel + Simulate	419	–	*DU*
HEU	Steel + Simulate	392	–	*LEU*
HEU	Steel + Simulate	157	–	HEU
HEU	Steel + Simulate	899	–	*LEU*
HEU	Lead	322	–	*LEU*
HEU	Lead	59771	*nat.U, elev. U conc.*	*LEU*

Table 4: Comparison of Detective EX analysis results for the uranium measurements yielded by the implemented analysis routine and calculated with FRAM; color code: red color refers to a completely incorrect identification result or no result at all, blue color refers to a correctly identified isotope or material, but the degree of enrichment or categorization is wrong, green color means both the identification and categorization are correct.

Sample	Shielding	Life Time (s)	Implemented Analysis	Result Derived From FRAM
DU	Unshielded	299	DU	DU
DU	Steel + Simulate	599	-	DU
DU	Steel + Simulate	937	DU	DU
Nat. U	Unshielded	201	*DU*	*DU*
Nat. U	Unshielded	89	*DU*	*DU*
Nat. U	Unshielded	103	*DU*	*LEU*

Sample	Shielding	Life Time (s)	Implemented Analysis	Result Derived From FRAM
Nat. U	Steel + Simulate	482	*DU*	*DU*
LEU	Unshielded	797	LEU, *WGPu*	LEU
LEU	Steel + Simulate	300	*DU*	*DU*
LEU	Steel + Simulate	221	*Nat. U*	LEU
LEU	Steel + Simulate	6199	*Nat. U*	LEU
LEU	Steel + Simulate	600	*Nat. U*	LEU
LEU	Lead	180	*DU*	*DU*
LEU	Lead	446	*DU*	LEU
HEU	Unshielded	264	*LEU*, WGPu	-
HEU	Unshielded	902	*LEU*, WGPu	HEU
HEU	Steel + Simulate	303	*LEU*	HEU
HEU	Steel + Simulate	617	*LEU*	*LEU*
HEU	Steel + Simulate	901	*LEU*	HEU
HEU	Steel + Simulate	22	*LEU*	*LEU*
HEU	Lead	179	*LEU*	-
HEU	Lead	58302	HEU, WGPu	HEU

Table 5: Comparison of FALCON 5000 analysis results for the uranium measurements yielded by the implemented analysis routine and calculated with FRAM; color code: red color refers to a completely incorrect identification result or no result at all, blue color refers to a correctly identified isotope or material, but the degree of enrichment or categorization is wrong, green color means both the identification and categorization are correct.

The Detective 200's implemented analysis routine does not include discrimination between LEU and HEU (merely "nuclear uranium" and HEU are defined) or between RGPu and WGPu (only "nuclear plutonium" is defined), respectively. Since the Micro Detective's routine is of the same type, no analysis results of this device are presented here as they were equally limited and therefore less interesting than the results by the Detective EX and the FALCON 5000, at least regarding the implemented routines.

In general, an obvious enhancement of the initial analysis results provided by the implemented routines was achieved with the FRAM analysis. The Detective 200 results benefitted most from the FRAM analysis as the latter produced the correct categorization in almost all cases. As for the other detectors, the FRAM analysis managed to enhance the previous identification result in some cases and failed to achieve this in others.

The use of FRAM appears to be most reasonable for the detector with the highest efficiency. The detectors mentioned above all cover a large range of gamma energy in order to analyse all types of radioactive and nuclear material. For cases when the focus of detection and identification is laid on nuclear material which emits photons in the lower energy region, specific U-Pu detectors are suitable. To evaluate if the FRAM analysis could be even more useful for a U-Pu germanium detector, a new series of measurements was done at INT with such a detector. Moreover, if the flaws of the detectors' implemented analysis routines were due to the large range of their gamma energy spectra and therefore due to limits of the energy resolution, these routines should be able to yield better results with spectra focussing on the lower energy region.

3.2 INT Measurements

The gamma energy spectra were collected by the detectors with the samples mentioned above. Tables 6 and 7 show, as examples, the comparison of the calculated mass percentages of ^{235}U in the sample of depleted uranium and the mineral sample, respectively, measured with the Detective 200, the Micro Detective, and the U-Pu detector, with 5 mm Fe shielding and without shielding.

Detector	No Shielding		5 mm Fe Shielding	
	Life Time (s)	^{235}U Content (%)	Life Time (s)	^{235}U Content (%)
Detective 200	26751	0.49 ± 0.01	6752	0.40 ± 0.03
Micro Detective	7005	0.16 ± 0.02	7030	0.17 ± 0.03
U-Pu Detector	7200	0.45 ± 0.06	7200	0.3 ± 0.1

Table 6: Mass percentages of ^{235}U in the sample of depleted uranium for three detectors as calculated by FRAM.

Detector	No Shielding		5 mm Fe Shielding	
	Life Time (s)	^{235}U Content (%)	Life Time (s)	^{235}U Content (%)
Detective 200	6752	1.0 ± 0.2	7200	0.0001 ± 0.0003
	28800	1.5 ± 0.1		
Micro Detective	3600	0.6 ± 0.3	28800	0.30 ± 0.06
	28800	0.6 ± 0.1		
U-Pu Detector	7200	3.2 ± 0.4	28800	2.4 ± 0.4

Table 7: Mass percentages of ^{235}U in the mineral sample for three detectors as calculated by FRAM.

The correct result of the ^{235}U mass percentage of the used depleted uranium would be approximately 0.25 % to 0.3 %. The Detective 200 and Micro Detective results were out of this range, even when taking the error figures into account. The results of the measurements with shielding, though, were closer to the expected value. The results for the U-Pu detector in general were higher than 0.3 %. The lower limit for the characterization as depleted uranium in general is ^{235}U < 0.64 %. This is fulfilled for all results, so an analysis would lead to the correct categorization.

For natural uranium the results were not in the correct range (0.64 % < ^{235}U < 0.78 %). In the shielded case the analysis was even more complicate. While the results for natural uranium are not correct this may not be of high concern for the decision makers. They are interested in correct results in terms of HEU/LEU in the case of uranium and of WGPu in the case of plutonium.

The fact that the FRAM analysis yielded not completely satisfactory results was primarily because of poor statistics leading to small numbers of counts in the relevant photo peaks. The samples used here emitted too weak a gamma radiation for an appropriate analysis. The FRAM analysis software was mainly designed for calculating enrichment of LEU or HEU samples and not for samples with such a low content of ^{235}U. In order to gain insight regarding the reasons for the flaws of the implemented analysis routines mentioned above by measurements with U-Pu detectors, other samples of nuclear material will be necessary. Therefore further measurements at institutions such as ITU must be taken into account.

4 Conclusions

Comparing the identification and categorization results of the detectors' implemented analysis routines to those achieved by means of the FRAM analysis with respect to measurements on uranium at ITU, a significant enhancement can be stated when using FRAM. In most cases, the implemented routines managed to identify the correct material, but the categorization was often false. Here the FRAM results were considerably superior, especially in the case of the Detective 200.

The measurements at ITU raised the question whether the flawed results obtained by the detectors' implemented analysis routines were due to limits of the measured spectra's energy resolution or because of weaknesses of the analysis routines themselves. To gain insight here, the additional measurements at INT with a U-Pu detector were performed. Unfortunately, the spectra measured with depleted uranium suffered from poor statistics due to the small amount of ^{235}U in the samples. Hence, other measurements with a U-Pu detector with low and high enriched uranium would be necessary to clarify this matter.

In general the quality of the spectra with electrically cooled germanium detectors was comparable to LN cooled germanium detectors. Therefore there is also a potential to enhance automatic analysis routines. Currently, the use of automatic analysis routines could be recommended only for unshielded or weekly shielded samples of SNM combined with a cautious review of the results.

5 References

[1] Risse, M.; Berky, W.; Friedrich, H.; Köble, T.; Rosenstock, W.; Rennhofer, H.; Pedersen, B.: *Identification of Nuclear Material with Hand-Held Gamma and Neutron Measuring Devices, INMM 51st Annual Meeting, Madison, Wisconsin: Omnipress, 2010, 10 S.*

[2] Risse, M.; Berky, W.; Friedrich, H.; Köble, T.; Rosenstock, W.; Schumann; O. Berndt, R.: *Identification Measurements of nuclear material – Detective EX versus Falcon 5000, INMM 53rd Annual Meeting, Orlando, 2012*

[3] Advanced Measurement Technology, http://www.ortec-online.com/download/PC-FRAM.pdf, downloaded on the 6th of July 2013

[4] Rosenstock, W.; Köble, T.; Risse, M.; *Measurement techniques to combat nuclear terrorism*; *Proceedings of the 25st annual meeting symposium on safeguards and nuclear material management*; Stockholm; 2003; 465