Contents lists available at ScienceDirect

# Applied Radiation and Isotopes

journal homepage: www.elsevier.com/locate/apradiso

# The Sandwich spectrometer for ultra low-level $\gamma$ -ray spectrometry

J.S. Elisabeth Wieslander<sup>a,d</sup>, Mikael Hult<sup>a,\*</sup>, Joël Gasparro<sup>a</sup>, Gerd Marissens<sup>a</sup>, Marcin Misiaszek<sup>b</sup>, Werner Preusse<sup>a,c</sup>

<sup>a</sup> EC-JRC-IRMM, Institute for Reference Materials and Measurements, Retieseweg 111, B-2440 Geel, Belgium

<sup>b</sup> M. Smoluchowski Institute of Physics, Jagiellonian University, ul. Reymonta 4, 30-059 Krakow, Poland

<sup>c</sup> Saxon State Laboratories for Environmental Radioactivity, Dresdner Street 183, D-09131 Chemnitz, Germany

<sup>d</sup> Department of Physics, P.O. Box 35 (YFL), FIN-40014 University of Jyväskylä, Finland

## ARTICLE INFO

Keywords: Ultra low-level gamma-ray spectrometry Muon Underground laboratory HPGe detectors Low background measurements

## ABSTRACT

The technical details and performance of the newly developed Sandwich spectrometer for ultra lowlevel  $\gamma$ -ray spectrometry are presented. The spectrometer, which consists of two HPGe detectors, an active muon shield and a lead/copper shield with a convenient and rapid opening mechanism, is located in an underground laboratory at a depth of 500 m water equivalent. The data is collected in list mode, which enables off-line data analysis to identify muon-induced events and possible Ge detector crosstalk due to Compton scattering. The background count-rate from 40 to 2700 keV normalised to the mass of the Ge crystals is 220 day<sup>-1</sup> kg<sup>-1</sup>.

© 2009 Elsevier Ltd. All rights reserved.

Applied Radiation and

# 1. Introduction

In the quest for lower detection limits in  $\gamma$ -ray spectrometry (Hult, 2007), a new spectrometer for ultra low-level gamma-ray spectrometry (ULGS) has been developed and characterised in an underground laboratory. Since the detection limit in  $\gamma$ -ray spectrometry is inversely proportional to the detection efficiency, it is of interest to design low-level detection systems with high efficiency. The detector presented here includes two Ge detectors facing each other between which the sample is placed: this approach effectively doubles the detection efficiency compared to using a single Ge detector. The new spectrometer design is intended for measurements of levels of activity in the mBq to  $\mu$ Bq range in small samples. The high efficiency allows faster measurements, which is suitable for a wide range of applications.

# 2. Properties of the Sandwich spectrometer

The sandwich spectrometer is located in the underground laboratory HADES (Hult et al., 2006, 2003) at a depth of 500 m water equivalent and located at the premises of the Belgian nuclear Centre SCK-CEN in Mol, Belgium. Fig. 1 shows a side and a top view of the Sandwich spectrometer while the electronics are described in Fig. 2.

During the design process for the Sandwich spectrometer extra attention was paid to the following points that affect system performance:

- Use of radiopure materials for the detectors and shielding.
- Radon reduction by minimising the empty cavities inside the shield and by flushing with liquid nitrogen boil-off from the two dewars.
- Easy access for changing samples as well as installing/ removing detectors.

## 2.1. HPGe detectors

The technical details of the two coaxial p-type HPGe detectors in Fig. 1, Ge-6 and Ge-7, are listed in Table 1. The samples are placed on Ge-6, which is always in a fixed position. Ge-7 can be moved vertically to accommodate bigger or smaller samples, which minimises not only the distance to the sample for both detectors but also the amount of air inside the measurement volume.

The relatively thick dead layer of Ge-6 in combination with the Cu endcap has the advantage of lower background and summing effects from low energy X-rays. The benefit of the Al endcap of Ge-7 in combination with the thin dead layer of its crystal is generally improved detection limits for  $\gamma$ -rays below 100 keV. Both detectors were well calibrated since they had been in use in the underground laboratory before they were installed in the Sandwich spectrometer, for which they were made available. Due to the multifunctional design of the lead shield, the Ge-detectors can easily be exchanged if necessary.



<sup>\*</sup> Corresponding author. Tel.: +32 14 571 269; fax: +32 14 584 273.

*E-mail addresses*: elisabeth.wieslander@gmail.com (J.S. Elisabeth Wieslander), mikael.hult@ec.europa.eu (M. Hult).

<sup>0969-8043/\$ -</sup> see front matter © 2009 Elsevier Ltd. All rights reserved. doi:10.1016/j.apradiso.2009.01.026



Fig. 1. The Sandwich spectrometer set-up including dimensions: (a) side view and (b) top view.



Fig. 2. The Sandwich spectrometer's electronics set-up showing how the two plastic scintillators, PS3 and PS4, and the two HPGe detectors, Ge-6 and Ge-7, are connected to the DAQ2000 multi-parameter system.

 Table 1

 Details of the two HPGe detectors included in the sandwich spectrometer set-up.

Details	Ge-6	Ge-7
Diameter × height (cm) Detector relative efficiency (%) FWHM at 1332 keV (keV) Dead layer (front) End-cap material Bias voltage (V) Year of manufacturing/refurbished	7.80 × 8.40 80 2.3 0.9 mm Cu +3000 2004	$\begin{array}{c} 8.05 \times 6.65 \\ 90 \\ 2.2 \\ 0.3  \mu m \\ Al \\ +4500 \\ 2004/2007 \end{array}$

## 2.2. Lead shield with Cu lining

The lead for the Sandwich spectrometer was produced by the Polish company Plombom and cast in the right shape by the company Von Gahlen. The innermost part of the lead shield is 4.0 cm thick with an activity of 2.5 Bq/kg (<sup>210</sup>Pb) while the outer part of the shield is 14.5 cm thick with an activity of 20 Bq/kg (<sup>210</sup>Pb).

To achieve lower background there is a 3.5 cm thick lining of radiopure electrolytic Cu closest to the measurement volume, within the lead shield, that shields against radiation from <sup>210</sup>Pb in the lead. The activity of <sup>60</sup>Co in freshly produced electrolytic copper could not be detected in a small test sample, so an estimate based on the fact that the copper was stored above ground for less than three weeks indicates that the <sup>60</sup>Co activity should be less than 15  $\mu$ Bq/kg (Laubenstein and Heusser, 2009). The activity of <sup>228</sup>Th in the electrolytic copper is estimated to be less than 20  $\mu$ Bq/kg.

The lead shield splits in two when it is opened, see Fig. 1b, and the front part is motor-driven along a rail which gives the advantage of making it fast and easy to open the shield to access samples and detectors. The diameter of the sample volume is slightly bigger than the diameter of the endcaps of the two Ge detectors, which is 102 mm. The upper Ge-detector (Ge-7) can be vertically moved in steps of 0.1 mm, resulting in a sample cavity height between 0 and 70 mm.

## 2.3. Active muon shield

Directly on top of the lead shield are two plastic scintillators (PS) as depicted in Fig. 1a and with details as listed in Table 2. The detectors are 2.54 cm thick and cover an area of  $80 \times 80$  cm<sup>2</sup>. Fig. 2 shows all Sandwich spectrometer electronics, including the coincidence circuit used for the hardware muon-gating pulse and the connection to the DAQ2000 multi-parameter system. The scintillator detectors are not shielded, which inevitably results in accidental coincidences originating from the environmental  $\gamma$ -background. Compared to the energy deposited by the muons, the  $\gamma$ -background is located in the low energy part of the spectrum and the hardware coincidence criteria reduce the background as seen in Fig. 3. The low-level discriminator (LLD) on the ADC is set high enough to filter out most of the  $\gamma$ -background but low enough to clearly see the valley between the muon and gamma background parts of the spectrum. The final muon signal is obtained by cutting off the  $\gamma$ -background at channel 840 in Fig. 3 during the off-line data analysis. Muons depositing high energy in the detectors cause saturation of the amplifier output and these are counted and collected at the end of the spectrum.

## 2.4. Electronics

The electronics included in the Sandwich spectrometer, see Fig. 2, consist of standard NIM modules together with the

### Table 2

Details of the plastic scintillators from Scionix, PS3 and PS4, included in the active muon shield.

Details	PS 3	PS 4	
Model number	R25 × 800 B 800/2 M–X+VD14–E2 with built-in Voltage divider/preamplifier		
Size (mm)	$800\times800\times25.4$		
Plastic type	EJ-200: PVT (polyvinyltoluene)		
Rise/decay time (ns)	0.9/2.1		
Photomultiplier tube	Type 2" ETL 9266, diameter 51 mm, 14 pins connector, Al housing		
Maximum signal height (Volt)	±10		
Spectrum sampled	No	Yes	
PM high voltage (V)	+1000	+1000	
Coincidence sampling	The coincidence signal between PS3 and PS4 is used as gating pulse to sample the muon spectrum from PS4		



Fig. 3. The muon energy spectrum in HADES using the coincidence signal from PS3 and PS4 as gate for the spectrum from PS4.

DAQ2000 multi-parameter system. The DAQ2000, which is based on LabView and designed and manufactured by IRMM, registers up to four input channels and time-stamps all events with 100 ns time step.

The coincidence signal from the two PS is used as a hardware gating signal *only* for the muon spectrum from the lower PS, PS4. *The coincidence circuit ensures that only gated signals are sampled from PS4 while all data from the two Ge detectors is collected.* For every event in any of the three detectors, Ge-6, Ge-7 or PS4, the time stamp is registered and the signal from all three detectors is collected. There is no hardware delay ensuring that the signals from the Ge detectors and the PS arrive at the same time. This is not needed because the coincidence criterion is set in the software during off-line analysis.

## 3. Data analysis and background reduction

The output from the DAQ2000 multi-parameter system consists of files with time-stamped list-mode data in binary format. The list files are converted to CERN standard for ROOT files (CERN, 2008) and the data is analysed using a custom-made software tool based on the ROOT environment. Since all events are collected and time-stamped, there are many choices for the off-line data analysis. The software is used for the usual  $\gamma$ -ray measurement analysis, for summing spectra from many measurements, performing energy calibration, studying time-interval distributions and finding coincidences or anti-coincidences between detectors for any chosen time window. Using the software, the coincidences with the muons are filtered out from the Ge detectors spectra, i.e. the muon-induced events are

#### Table 3

List of background  $\gamma$ -ray peaks in the sandwich detector with the uncertainties in brackets. The last rows show the count-rate in the background for different energy ranges, with the number of derived muons in brackets.

E <sub>γ</sub> (keV)	Radionuclide	Ge-7 without muons (counts day <sup>-1</sup> )	Ge-6 without muons (counts day <sup>-1</sup> )
46	<sup>210</sup> Pb	2.1 (3)	<0.5
63	<sup>234</sup> Th	< 0.6	< 0.6
93	<sup>234</sup> Th	< 0.6	< 0.6
186	<sup>226</sup> Ra+ <sup>235</sup> U	< 0.7	< 0.7
238	<sup>212</sup> Pb	2.2 (3)	2.1 (4)
242	<sup>214</sup> Pb	0.34 (25)	< 0.8
295	<sup>214</sup> Pb	0.70 (29)	0.56 (26)
338	<sup>228</sup> Ac	0.28 (17)	< 0.6
351	<sup>214</sup> Pb	2.1 (3)	2.3 (3)
511	Annihilation	2.51 (28)	2.9 (4)
583	<sup>208</sup> Tl	1.05 (19)	0.61 (19)
609	<sup>214</sup> Bi	2.37 (25)	2.5 (0.3)
661	<sup>137</sup> Cs	< 0.3	< 0.4
911	<sup>228</sup> Ac	0.48 (14)	0.79 (20)
969	<sup>228</sup> Ac	0.22 (11)	0.26 (14)
1120	<sup>214</sup> Bi	0.50 (13)	0.86 (19)
1173	<sup>60</sup> Co	1.44 (17)	0.85 (18)
1332	<sup>60</sup> Co	1.24 (16)	1.32 (20)
1460	<sup>40</sup> K	1.31 (16)	0.84 (17)
1764	<sup>214</sup> Bi	0.41 (10)	0.72 (14)
	40-2400 keV	428 (72 muons)	441 (52 muons)
	40-400 keV	264 (35 muons)	268 (39 muons)
	400-1400 keV	136 (28 muons)	145 (19 muons)
	1400-2400 keV	28 (9 muons)	28 (5 muons)
	Measurement	46.4 days	63.4 days
	time		

removed. The time window between the arrival of the signal from the PS to the arrival of the slower pulse from the Ge detectors used for these anti-coincidences can be selected freely. In this work it is taken to be 1 ms. The crosstalk due to Compton scattering between the two Ge detectors can also be reduced with software analysis.

## 3.1. Uncertainties

The uncertainties are expressed as combined standard uncertainties following the BIPM Guide to the Expression of Uncertainty in Measurement (1995) while detection limits and decision thresholds are in accordance with ISO 11929-3 (2000), see Table 3. All results are compared to the decision threshold using  $\alpha = 0.05$ . The uncertainty is given in brackets after each measurement value and the last digit(s) of their numerical value correspond(s) to the last digit(s) of the quoted result. The major contribution to the uncertainty budget in this work originates from counting statistics. The reference date and measurement times must be precisely recorded.

# 4. The Sandwich spectrometer performance

## 4.1. Solid angle

The maximum solid angle for the Ge detectors in the Sandwich spectrometer has been calculated to 3.0 sr (corresponding to 24% of  $4\pi$ ) using the following approximations:

- The end-caps of the two detectors are so close that they touch each other.
- An infinitesimally thin sample of 1 cm in diameter is placed between the end-caps, i.e. in contact with each detector's end-cap.

• The dead layers are excluded in the calculation of the solid angle and only the active crystal volume is included.

# 4.2. Efficiency

Generally, the full energy peak (FEP) efficiency for a given sample is a factor of two higher for the Sandwich spectrometer as compared to a single Ge detector. For examples see the decay data measurements of a thick tantalum metal disc given by Hult et al. (2009).

## 4.3. Muons in HADES

The average count-rate of muons in the Sandwich spectrometer active muon shield, see Fig. 3, is  $0.138 \text{ muons m}^{-2} \text{s}^{-1}$ during the period July 2007 to May 2008. The count-rates of muon-induced events in the two Ge detectors are 72 muons day<sup>-1</sup> and 52 muons day<sup>-1</sup> for Ge-6 and Ge-7, respectively.

## 4.4. Background reduction with the active muon shield

In Fig. 4 the background spectrum for the Sandwich spectrometer is shown without the muon-induced events. The events that were filtered out are shown in Fig. 5. The background count-rates of the Sandwich spectrometer in the energy interval 40–2400 keV are 992 counts day<sup>-1</sup> with the muons included and 868 counts day<sup>-1</sup> without muons, the muons being 124 counts day<sup>-1</sup>, see Table 3. Extrapolating the background from 2400 to 2700 keV enables comparison with other underground spectrometers. The count-rate



Fig. 4. The background  $\gamma$ -ray energy spectrum from the Sandwich spectrometer with the Ge detectors Ge-6 and Ge-7 summed together and the muon-induced events filtered out.





normalised to the Ge-crystal mass in the region 40-2700 keV is  $220 \text{ day}^{-1} \text{ kg}_{(Ge)}^{-1}$ , which is lower than the previous best detector in the HADES laboratory: Ge-3 with a background index of 280 day<sup>-1</sup>  $kg_{(Ge)}^{-1}$  During the measurement of a tantalum sample with a diameter of 100 mm (Hult et al., 2009) the count-rates were reduced by about 10% compared to the background, especially below 500 keV due to the shielding effect of the tantalum sample that was 12 mm thick. Generally the background count-rates from the two detectors are very similar, the major contribution being the <sup>222</sup>Rn daughters. It is likely that the main source of the <sup>222</sup>Rn daughters is <sup>226</sup>Ra inside the detectors. The 186 keV peak from <sup>226</sup>Ra is below the detection limits in the background measurements but can be seen in the sum spectrum from the tantalum measurements (Hult et al., 2009), which leads to the conclusion that the contribution to the background from <sup>222</sup>Rn transported via air from outside the shield is not a major factor.

# 5. Discussion

The new Sandwich spectrometer with its high efficiency and low background is ideal for many applications involving small samples or radionuclides with relatively short half-lives. In several applications it is necessary to measure activation products in small metal samples, such as: (i) neutron cross-section measurements using the traditional activation approach (Reimer et al., 2002), (ii) neutron cross-section curve measurements using broad beams (Lövestam et al., 2007), (iii) fast neutron dosimetry and spectrum characterisation (Lövestam et al., 2008), (iv) samples activated by charged particle leakage from fusion plasma (Wieslander et al., 2008) and decay measurements of rare decays such as in  $^{180m}$ Ta (Hult et al., 2009) or the  $\beta\beta$ -decay in Sn (Dawson et al., 2008). The Sandwich spectrometer is also an efficient way of using resources, since less lead and copper is needed in comparison to two single systems. Based on the experiences from this spectrometer, the next generation of HADES Sandwich spectrometer is being designed. The new system has a larger sample cavity, for which it is necessary to further develop the active radon-reduction inside the shield. In addition to enabling measurements of larger volume samples, a larger sample cavity would also open up the possibility of placing other types of detectors, such as small scintillator cells, inside the shield for special coincidence or anti-coincidence measurements.

## Acknowledgements

The work done by EURIDICE and the HADES crew of SCK • CEN in Mol, Belgium, is gratefully acknowledged.

### References

- CERN, 2008. <www.root.cern.ch/>, accessed 2008.
- Dawson, J., Degering, D., Köhler, M., Ramaswamy, R., Reeve, C., Wilson, J.R., Zuber, K., 2008. Search for double beta decays of tin isotopes with enhanced sensitivity. Physical Review C 78 (3), 035503.
- Hult, M., 2007. Low-level gamma-ray spectrometry using Ge-detectors. Metrologia 44, S87–S94 (Erratum in Metrologia 44, 425).
- Hult, M., Gasparro, J., Johansson, L., Johnston, P.N., Vasselli, R., 2003. Ultra sensitive measurements of gamma-ray emitting radionuclides using HPGe-detectors in the underground laboratory HADES. Environmental Radiochemical Analysis, vol. II, Royal Society of Chemistry, Cambridge, UK, pp. 375–382.
- Hult, M., Preuße, W., Gasparro, J., 2006. Underground Gamma-ray spectrometry. Acta Chimica Slovenica 53, 1–7.
- Hult, M., Wieslander, J.S.E., Marissens, G., Gasparro, J., Wätjen, U., Misiaszek, M., 2009. Search for the radioactivity of <sup>180m</sup>Ta using an underground HPGe Sandwich spectrometer. Applied Radiation and Isotopes, these proceedings, doi:10.1016/j.apradiso.2009.01.057.
- ISO/IEC/OIML/BIPM, 1995. Guide to the Expression of Uncertainty in Measurement, first ed., International Standard Organisation, Geneva, Switzerland.
- ISO, 11929-3, 2000. Determination of the Detection Limit and Decision Threshold for Ionising Radiation Measurements—Part 3, first ed., International Standards Organisation, Geneva, Switzerland.
- Laubenstein, M., Heusser, G., 2009. Cosmogenic radionuclides in metals as indicator for sea level exposure. Applied Radiation and Isotopes, these proceedings, doi:10.1016/j.apradiso.2009.01.029.
- Lövestam, G., Hult, M., Fessler, A., Gamboni, T., Gasparro, J., Geerts, W., Jaime, R., Lindahl, P., Oberstedt, S., Tagziria, H., 2007. A novel technique for measuring neutron activation cross section curves. Nuclear Instruments and Methods A 580, 1400–1409.
- Lövestam, G., Hult, M., Fessler, A., Gasparro, J., Kockerols, P., Okkinga, K., Tagziria, H., Vanhavere, F., Wieslander, J.S.E. Neutron fluence spectrometry using disk activation. Radiation Measurements, in press, doi:10.1016/j.apradiso. 2008.10.012.
- Reimer, P., Hult, M., Plompen, A.J.M., Johnston, P.N., Avrigeanu, V., Qaim, S.M., 2002. Measurement of the <sup>nat</sup>Mo(n,x)<sup>94</sup>Nb cross section using ultra low-level γ-ray spectrometry at HADES. Nuclear Physics A 705, 265–278.
- Wieslander, J.S.E., Hult, M., Bonheure, G., Arnold, D., Dombrowski, H., Gasparro, J., Laubenstein, M., Marissens, G., Vermaercke, P., 2008. Low-level gamma-ray spectrometry for analysing fusion plasma conditions. Nuclear Instruments and Methods A 591, 383–393.