STATUS OF UNDERGROUND RADIOACTIVITY MEASUREMENTS IN HADES

E. Andreotti[1], M. Hult[1], R. Gonzalez de Orduña[1], G. Marissens[1], M. Mihailescu[1], U. Wätjen[1], P. Van Marcke[2]

[1] *EC-JRC Institute for Reference Materials and Measurements, Geel, Belgium*
[2] *The Belgian Nuclear Research Centre SCK•CEN, Mol, Belgium*

The IRMM (Institute for Reference materials and Measurements) performs ultra low-level gamma-ray spectrometry at a depth of 225 m in the underground laboratory HADES. The facility currently houses 7 HPGe-detectors that are built and shielded using specially selected radiopure materials. The sand-clay overburden of about 500 m water equivalent assures a muon flux reduction factor of about 5000, with subsequent reduction of the background of the detectors, which makes it possible to obtain detection limits close to 100 μBq for certain radionuclides. This paper describes the aim of the IRMM activities in the HADES laboratory, the equipment and the measurement program and gives examples of radiopurity measurements carried out in order to develop better low-level measurements.

1. Introduction

The field of nuclear physics experiments in underground laboratories has expanded significantly in recent years. The main developments and investments are in the field of large scale astroparticle physics experiments with detectors like SNO, KamLAND, OPERA, Borexino, GERDA, CUORE etc. [1]. Parallel to this development, underground measurements of radioactivity using laboratory work-horses like HPGe-detectors, have increased. This increase is triggered both by the need of the large scale experiments to use radiopure materials, as well as emerging fields where underground radioactivity measurements can provide significant improvements.

The Institute for Reference Materials and Measurements (IRMM)[1], sited in Geel (Belgium), is a part of the European Commission's Joint Research Centre (JRC) and has as mission to promote a common and reliable European measurement system in support of EU policies. IRMM began its underground research in collaboration with its neighbour, the Belgian nuclear centre SCK•CEN[2], in 1992 with only one detector taking advantage of the HADES underground laboratory located on the premises of SCK•CEN. In the beginning this was an exploratory activity, but relatively quickly it became clear that underground gamma-ray spectrometry could be used to give an extra edge in many fields of research in which IRMM is involved. This has lead to an expansion of the activity and the laboratory hosts 7 HPGe-detectors at present. The strategy of IRMM's underground work has been to provide special measurements within the fields of priority of the institute, which include e.g. nuclear safety, metrology and nuclear decay data.

The present fields of competence of IRMM in the nuclear field can partly be traced back to its historical origin, which dates back to the 1960ies. The institute was founded in 1957 under the EURATOM Treaty and started operation in 1960 under the name of Central Bureau for Nuclear Measurements (CBNM). In 1993 CBNM was renamed in order to reflect the enlarged frame of competences, covering a wide range of additional measurement problems, related to food safety and chemical metrology. At present IRMM assumes a key role in the organization of regular measurement comparisons among EU national metrology as well as monitoring laboratories and is one of the world's leading producers of reference materials.

The HADES (High Activity Disposal Experimental Site) underground laboratory, hosting the IRMM's Ultra Low-level Gamma-ray Spectrometry (ULGS) facility, was built in the frame of the Belgian research programme regarding the geological disposal of radioactive waste, on the premises of the Belgian Nuclear Research Centre SCK•CEN in the town of Mol. Since 1997 the HADES underground laboratory is managed by EURIDICE[3] (European Underground Research Infrastructure for Disposal of nuclear waste In Clay Environment). HADES is a research facility and not a disposal site. Hence there are no plans to place radioactive waste in it.

The main reason for performing gamma-ray spectrometry underground is the significant reduction of the background due to the strong attenuation of the cosmic ray induced muon flux. In order to fully exploit the advantages of performing underground measurements it is essential to use radiopure materials for the construction of both the detector and the shielding. The search for rare events, like double beta decay or dark matter interaction, provides one of the principal driving forces to the development and improvement of underground science and to the progress in radiopure material selection. In connection to this the ILIAS[4] (Integrated Large Infrastructures for Astroparticle Science) project has been an important catalyst by providing co-ordination of activities and underground infrastructures for astroparticle physics at a European level, in the frame of the European Commission 6th Framework Programme (FP6). Since IRMM joined ILIAS its ULGS facility has been involved in several activities connected to the

---

[1] IRMM website: http://irmm.jrc.ec.europa.eu/

[2] SCK•CEN (StudieCentrum voor Kerenergie•Centre d'Etude Nucleaire) website: http://www.sckcen.be/en/

[3] EURIDICE is an Economic Interest Group (EIG) between SCK-CEN and NIRAS/ONDRAF, the Belgian Agency for Radioactive Waste and Enriched Fissile Materials.

[4] ILIAS website: http://www-ilias.cea.fr/

improvement of underground measurement techniques. The IRMM's ULGS laboratory is furthermore involved in CELLAR (Collaboration of European Low-level underground Laboratories), a network created in the year 2000, in order to promote underground radioactivity and dosimetry measurements [2].

2. The underground laboratory HADES

In Belgium, the R&D programme for the long-term management of long lived High Level Waste (HLW) was initiated at the Belgian nuclear research centre (SCK•CEN) in 1974. The so-called "Boom Clay" was selected as a potential host formation for the disposal of the HLW. Preliminary laboratory research yielded promising results, thus it was decided to construct the underground research facility HADES (High-Activity Disposal Experimental Site) at 225 m depth. Fig. 1 shows the layout and construction history of the laboratory.

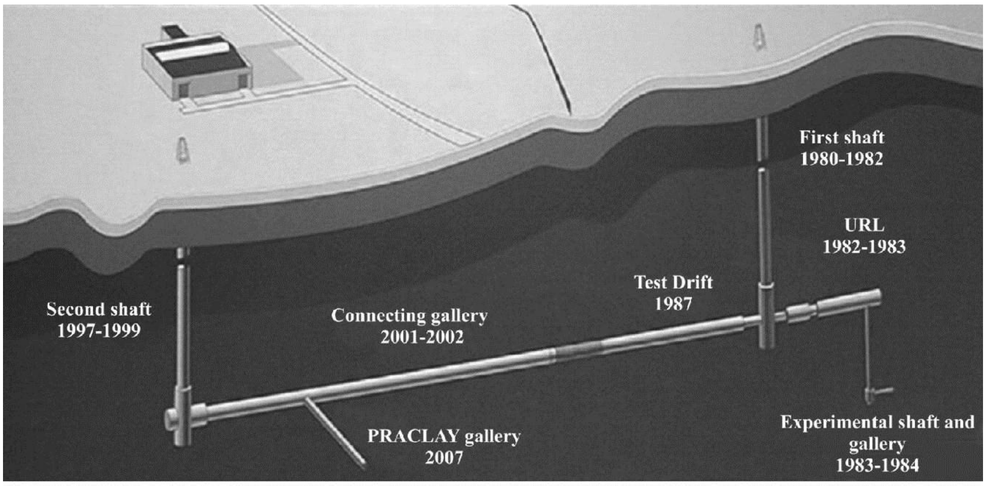

Fig. 1. Layout and construction history of the underground laboratory HADES

The main purpose of the laboratory was to examine the possibility of constructing a repository in this type of clay at such a depth and to conduct various in-situ experiments with the aim of characterizing geological properties of the clay. The Boom Clay was deposited in the Tertiary Period which lasted from 36 to 30 million years ago. It is found at a depth of about 190 metres under the site of Mol, where it has a thickness of about 100 metres. The clay layer is almost horizontal and water bearing sand layers are situated above and below it. It is a silty clay characterised by a structure of bands that are several tens of centimetres thick which are mainly caused by fluctuations in the wave action on the sedimentation medium. Due to this vertical heterogeneity there is also a wide variation in the content of clay minerals (from 30 to 70 % volume, dry matter) [3]. Due to the very low hydraulic conductivity of the clay (10^{-12} m/s) only a very small influx of water in the laboratory occurs, which is evaporated immediately.

The IRMM's ULGS facility is located in the part of the gallery called the Underground Research Laboratory (URL), which is 32 m long and has an inside diameter of 3.5 m. The lining of the laboratory is made of 50 mm thick cast iron segments. These are reinforced with 200 mm thick iron ribs that cover 25 % of the wall area.

2.1 Characterization of the underground site

The radioactivity of the clay and sand layers, which compose the overburden of the underground laboratory, was measured using gamma-ray spectrometry. The main components of the radioactivity in the clay comes from ^{238}U (80 Bq/kg), ^{232}Th (40 Bq/kg) and ^{40}K (700 Bq/kg), all activities in dry weight. This clay activity is thus close to the average of the earth's crust but less radioactive than some granites, albeit it is not as radiopure as salt generally is. However, the presence of the cast iron lining covering the walls of the URL, helps to shield from the background contributions due to the gamma emitters in the clay. The radioactivity of the cast iron lining (stored above ground) has also been measured and its main component is from the ^{232}Th decay chain. The massic activities of ^{228}Ra and ^{228}Th are 0.144 ± 0.010 and 0.174 ± 0.012 Bq/kg respectively. Considering that the lining has been kept in HADES for 18 years, the relatively short-lived cosmogenic radionuclides ^{57}Co, ^{58}Co, ^{54}Mn, ^{56}Mn, ^{59}Fe and ^{51}Cr have decayed to insignificant levels. No presence of ^{60}Co was detected and the decay corrected detection limit of ^{60}Co in the iron in HADES is 0.25 mBq/kg.

The dose rate in the laboratory was measured using two instruments: a plastic scintillator probe (Automess 6150 AD-b) and a NaI-probe (Canberra, Inspector 1000). The readings from the two instruments agreed well. The dose rate in the laboratory depends strongly on the lining. In the Connecting gallery (see Fig. 1) with a new concrete lining, the dose rate is 70 nSv/h. The first part of the Test Drift (see Fig. 1) has an iron rib lining where the dose rate is 40 nSv/h. The second part of the Test Drift has an old concrete lining and a dose rate of 47 nSv/h. Finally, in the oldest part (the URL in Fig. 1), where the gamma spectrometers are located, the dose rate is only 15 nSv/h due to the very thick cast iron lining. At certain places in the URL where there are extra thick iron plugs, the dose rate was as low as

7 nSv/h. A measurement was also carried out inside one of the lead/copper shields that are used for the HPGe-detectors, which gave a result below the detection limit of 5 nSv/h.

The radon concentration in air is constantly monitored using an AlphaGuard. Variations are registered during the year, with maximum values not exceeding 20 Bq/m^3 and an average value of about 5 Bq/m^3. This very low concentration for an underground site is attained thanks to the ventilation system operating in the laboratory, which generates a constant air flow of about 10.000 m^3/h.

The sand and clay overburden of 225 m (about 175 m sand and 50 m clay), corresponds to ~500 m w.e. and assures a muon flux reduction of about 4 orders of magnitude with respect to ground level (less than 0.2 m^{-2} s^{-1}). This estimation is based on measurements performed with the plastic scintillators covering the Sandwich spectrometer, described in section 3. The neutron fluence rate is less than 2 m^{-2} s^{-1} for both thermal and fast neutrons. This result was deduced during preliminary measurements from the neutron induced peaks produced by inelastic scattering and activation in one of the germanium detectors installed underground [4].

Together with the radon concentration in air, environmental conditions including temperature, humidity and air pressure are constantly monitored. The temperature is very stable around 25 °C with variations generally less than a few tenth of a degree. The humidity and pressure do, however, vary following the atmospheric conditions. The air pressure measured in HADES is 2.7 kPa higher than that measured above ground (~20 m above sea level).

3. The gamma-ray spectrometry laboratory

Fig. 2. View of the part of HADES hosting the IRMM gamma-ray spectrometry equipment.

Fig. 2 shows the part of HADES where the IRMM gamma-ray spectrometry equipment is located. The laboratory is not sited in a clean room and the environment is less clean than in a normal gamma spectrometry laboratory. This has however not affected the measurements since the lead shields are very tight (and covered by inflammable dust covers). The background of each HPGe-detector is regularly measured and it continues to decrease due to the decay of the cosmogenic radionuclides in the detector itself and in the shield. No increase in e.g. ^{210}Pb has been registered, which one could expect in case the dust posed a problem.

IRMM operates at present seven HPGe-detectors. All detectors are made from selected radiopure material and have special low-background features, as for example the location of the pre-amplifier far away from the crystal. The main characteristics of the present and planned detectors are listed in Table 1.

Table 1. **Overview of HPGe-detectors located in the HADES laboratory**

	Ge2[a]	Ge3	Ge4[a]	Ge5[a]	Ge6[a]	Ge7[a,b]	Ge8[a]	Ge9[c]	Ge10[d]
Status	In operation	In operation	In operation	In operation	In operation	In operation	In operation	Tests in progress	Ordering in progress
Detector type	n type semiplanar	p type coaxial	p type coaxial	p type planar	p type coaxial	p type coaxial	p type planar	p type planar	n-type, coaxial
Relative efficiency (%)	8	60	106	50	80	90	19	50	60*
Mass (kg)	0.202	1.269	2.113	0.804	2.134	1.802	0.391	0.84	1.3*
Window material	Al	Al	Al	Al	Cu	Al	Al	Al	Al
FWHM @ 1332 keV	1.60	1.99	1.99	1.81	2.15	2.08	1.72	1.62	2.2*
Installed (year)	1994	1996	1999	2001	2003	2005	2006	2010	2010*

Special features: a. submicron deadlayer; b. inverted endcap; c. Ge depleted in ^{76}Ge; d. optimised for Compton suppression. *Expected values.

Detectors 6 and 7 are placed face-to-face inside the same shielding, in order to improve the efficiency by increasing the solid angle. This spectrometer is called "Sandwich-spectrometer" and has the limitation of being able to measure only relatively small samples [5]. The Sandwich spectrometer is furthermore covered by a pair of large area plastic scintillators, operated in coincidence, which reduces the residual muon contribution to the background with about 30 %.

The installation of a new innovative system, the so-called "Pacman-spectrometer", is ongoing. The shield of the Pacman-spectrometer is larger than the sandwich and has a more flexible scope. It can host two HPGe-detectors facing each other (like "the Sandwich") or it can host one HPGe surrounded by a massive Compton suppression shield (NaI or BGO).

The shielding surrounding all ULGS detectors typically consists of 15 - 25 cm of lead of which the inner 2 - 5 cm are low in ^{210}Pb (< 3 Bq/kg), plus an inner lining of 1 - 15 cm of freshly produced electrolytic copper. Such kind of shielding is very well suited for underground measurements, where the muon flux is sufficiently reduced in order not to increase the background due to activation in copper and to secondary radiation originated by cosmic rays interacting in the shielding itself. The inner low radioactive lead was obtained from several hundred years old roof tiles, pipings and monuments in connection with refurbishments at Versailles, Hampton Court Palace and het Pand (in Ghent). The inner copper lining is made from electrolytic copper for which the time above ground has been minimised in order to keep the cosmogenic activation as low as possible. Radon reduction is achieved by minimising the empty space inside the shield and flushing with nitrogen evaporating from the liquid nitrogen Dewars.

The background count rate of the different ULGS detectors per kg of germanium does not vary a great deal. The typical background for a ULGS detector is in the order of 300 counts per day and kg of Ge in the energy region between 40 and 2700 keV. The best value at present is for the sandwich-spectrometer with 220 counts per day.

4. Measurement program

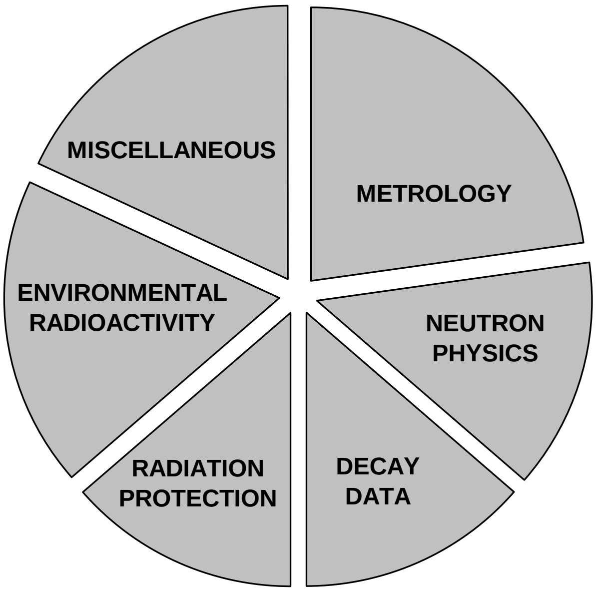

Fig. 3. HADES activities overview 2004 - 2009.

The measurement program is strongly governed by IRMM's mission of providing support to EU-policies and the scientific pillars of the IRMM. Fig. 3 shows an overview of how the measurement time on ULGS detectors in HADES was used during FP6 and the first two years of FP7, i.e. 2004 - 2009.

The fields Metrology [6], Neutron Physics [7] and Decay Data [8] have been key areas within the mission of IRMM since its foundation. The field radiation protection involves projects like the measurements of samples from Hiroshima for retrospective dosimetry [9]. Such measurements, combined with epidemiological studies of Hiroshima survivors, form the basis for the legislation on radiation protection. The field environmental radioactivity involves e.g. a study on the possibility of distinguishing organic farm products from conventional ones by measuring their radioactivity. The basic idea is that conventional fertilizers based on phosphate ore are higher in natural radioactivity than organic manure [10]. Improving the possibility of identifying such farm products (or at least raising a flag) would potentially reduce fraud in this field and increase consumers trust. Amongst measurements counted in the field miscellaneous measurements one finds support to large-scale underground experiments like GERDA [11].

4.1. Radiopurity measurements

Improving the background of low-level measurements is partly an iterative process. One needs to identify radiopure materials in order to construct better detectors with which one can identify even lower activities. A general problem with selecting materials is that there is a great variation in radiopurity between different batches of the same material. This calls for large numbers of measurements. Table 2 presents the activities measured in some materials important for ULGS where the radiopurity demand is high.

Carbon epoxy windows are commonly used on HPGe-detectors. The results in Table 2 show that these are not suitable for underground HPGe-detectors since the ^{40}K and ^{226}Ra (in Batch 2) activities are too high. In the past aluminium used to have too high levels of thorium for use in underground detectors. Today it is possible to purchase aluminium with a very high level of radiopurity (see Table 2) although still not as good as electrolytic copper. When using an aluminium window, it is best if the whole endcap is made from aluminium to avoid the problem of making a radiopure fixation between the window and the endcap.

Table 2. **Results of radiopurity measurements in IRMM's ULGS. The quoted uncertainties are combined standard uncertainties and the decision thresholds are calculated using an error of first kind (α) equal to 0.05**

Material	Mass, g	Measuring live time, d	^{238}U, Bq kg^{-1}	^{226}Ra, Bq kg^{-1}	^{228}Ra, Bq kg^{-1}	^{228}Th, Bq kg^{-1}	^{40}K, Bq kg^{-1}
Carbon Epoxy Batch-1	26	7	$< 7 \times 10^{-2}$	$< 5 \times 10^{-2}$	$< 5 \times 10^{-2}$	$< 5 \times 10^{-2}$	3.2 ± 0.4
Carbon Epoxy Batch-2	81	7	$< 5 \times 10^{-2}$	$(4.0 \pm 1.5)\times10^{-2}$	$< 3 \times 10^{-2}$	$< 3 \times 10^{-2}$	2.4 ± 0.5
Aluminium endcap-1	173	14	$< 5 \times 10^{-2}$	$< 9 \times 10^{-2}$	$< 2.5 \times 10^{-2}$	$< 2 \times 10^{-2}$	$< 1 \times 10^{-1}$
Aluminium endcap-2	260	26	$< 2 \times 10^{-2}$	$< 1.5 \times 10^{-2}$	$< 7 \times 10^{-3}$	$< 4 \times 10^{-3}$	$< 3 \times 10^{-2}$
Phenoseal	218	26	$(7.0\pm 1.3) \times10^{-2}$	$(3.1 \pm 0.3)\times10^{-2}$	$(1.4\pm 0.3)\times10^{-2}$	$(2.2 \pm 0.2)\times10^{-2}$	$(2\pm 0.4)\times10^{-1}$
TEC-7	479	2	3.5 ± 0.7	2.6 ± 0.2	$< 2 \times 10^{-1}$	$(3.6 \pm 0.6)\times10^{-1}$	$< 3 \times 10^{-1}$
Viton O-rings	77	24	$(1.5 \pm 0.5)\times10^{-1}$	$(4.4 \pm 0.3)\times10^{-1}$	$(6.7 \pm 1.5)\times10^{-2}$	$(8.0 \pm 1.5)\times10^{-2}$	2.7 ± 0.3

A common problem in underground radioactivity measurements is to seal containers. The reason could be that they contain liquids or that the radon daughters must be contained since they are used for quantifying ^{226}Ra. Liquids can be contained (although not safely sealed) using a container lid that snaps onto the container like many Marinelli beakers have. A more robust way is to close with screws and use an O-ring, but O-rings are generally not radiopure. Table 2 shows a measurement of a batch of Viton O-rings, which is relatively radiopure. An alternative sealing approach is to use glues or silicone based sealants. This has been investigated by e.g. Parekh et al. [12] and Busto et al. [13]. The former mentioned that phenoseal could be a suitable material and presented detection limits in the order 5-10 Bq/kg for ^{40}K, ^{210}Pb and ^{226}Ra. The results in Table 2 show that Phenoseal is more radiopure than most sealants but the radioactivity levels must be taken into account when measuring underground. Another glue, TEC-7, that is being used for above ground measurements, was also tested. It is clearly not suitable for underground work.

Acknowledgements

The HADES staff of EURIDICE is gratefully acknowledged for their work. Many thanks to Raf Aarts (SCK•CEN) for performing the dose rate measurements.

REFERENCES

1. *Strumia A., Vissani F.* Neutrino masses and mixing and … // arXiv:hep-ph/0606054v3. - 2006. - http://arxiv.org/abs/hep-ph/0606054v3.
2. *Laubenstein M., Hult M., Gasparro J. et al.* Underground measurements of radioactivity // Appl. Radiat. Isot. - 2004. - Vol. 61. - P. 167 - 172.
3. *Honty M.* SMARAGD: Study of Mineral Alterations of clay barriers used for Radwaste storage and its Geological Disposal // Final report. Project co-funded by the EC, Contract # FI6W-CT2005-028403. - DG for Research EURATOM, Luxembourg, 2008. - EUR 23468. - 70 p.
4. *Wordel R., Mouchel D., Bonne A.et al.* Low-level gamma-ray measurements in a 225 m deep underground laboratory // Proc. of the 3rd Int. Summer School (Huelva, Spain) / Ed. by M. Garcia-León, R. Garcia-Tenorio. - Singapore: World Scientific, 1993. - P. 141 - 153.
5. *Wieslander J.S.E., Hult M., Gasparro J. et al.* // The Sandwich spectrometer for ultra low-level γ-ray spectrometry // Appl. Radiat. Isot. - 2009. - Vol. 67. - P. 731 - 735.
6. *Hult M.* Low-level gamma-ray spectrometry using Ge-detectors // Metrologia. - 2007. - Vol. 44. - P. S87 - S94.
7. *Wieslander J.S.E., Lövestam G., Hult M. et al.* // Validation of a method for neutron dosimetry and spectrometry using neutron activation of metal discs / Rad. Prot. Dos. - 2010. - Vol. 138(3). - P. 205 - 212.
8. *Wieslander J.S.E., Suhonen J., Eronen T. et al.* Smallest known Q value of any nuclear decay: the rare β^- decay of ^{115}In($9/2^+$) → Sn($3/2^+$) // Phys. Rev. Lett. - 2009. - Vol. 103. - P. 122501.
9. *Gasparro J., Hult M., Marissens G. et al.* Measurements of ^{60}Co in massive steel samples exposed to the Hiroshima atomic bomb explosion // JRC scientific and technical reports. - European Communities, Luxembourg, 2010. - EUR 24146 EN. - 31 p.
10. *Lindahl P., Hult M., Maquet A. et al.* Natural radioactivity in winter wheat from organic and conventional agricultural systems // J. Env. Rad. - 2011. - Vol. 102(2). - P. 163 - 169.
11. *Janicskó-Csàthy J.* Status of the GERDA experiment // Nucl. Phys. B (Proc. Suppl.). - 2009. - Vol. 188. - P. 68 - 70.
12. *Parekh P.P., Bari A., Semkow T.M., Torres M.A.* A new method for sealing containers with liquid samples for radioactivity measurements // J. Rad. Nucl. Chem. - 2002. - Vol. 253(2). - P. 321 - 325.
13. *Busto J., Gonin Y., Hubert F. et al.* Radioactivity measurements of a large number of adhesives // Nucl. Inst. and Meth. A. - 2002. - Vol. 492. - P. 35 - 42.