Radionuclide Identifier for Gamma Analysis – RIGA

A. Rozite

European Commission Joint Research Centre, Ispra, Italy
E-mail: arturs.rozite@ec.europa.eu

Abstract

A hand-held HPGe spectrometer with a hybrid cooling system is developed. The cooling system of the spectrometer is liquid nitrogen-free and provides optimal energy resolution for a planar HPGe detector. The instrument is created for nuclear safeguards inspectors for such applications as U enrichment measurements and Pu isotopic composition verification with MGA, MGAU and FRAM software codes.
A cooling system of the instrument contains a cryogen, which is pre-cooled to the solid crystalline state by a detachable Stirling-cycle cryocooler. When cooled the cryogen works as a thermal buffer keeping the temperature of the detector low enough and stable using the energy barrier accumulated at the phase transition from the solid to the liquid state.
The cooling time of the cryogen from room to melting temperature is 7 hours, and the autonomous operation time of the device before the cryogen re-cooling is 4 hours.
The weight of the instrument carried by an operator during measurements is 3.9 kg, the dimensions are 364 × 92 × 239 mm. *The weight of the docking station based on the cryocooler is 4.5 kg.*
The name of the device is RIGA – Radionuclide Identifier for Gamma Analysis.
In the article thermal and vacuum properties of the cryostat, as well as mechanical and spectrometric characteristics of the device are presented.

Keywords: HPGe detector, nuclear safeguards, uranium, plutonium

1. Introduction

Semiconductor HPGe detectors are widely used for the measurements of radiation due to the advantage in the energy resolution they provide over scintillation ones, but to operate they need to be cooled to low, almost cryogenic temperatures.
Traditionally HPGe detectors have been cooled by liquid nitrogen, and compact HPGe spectrometers for in-situ applications utilizing this method of cooling have been developed in the past [1–3]. Later, to avoid inconveniences associated with the usage of liquid nitrogen, and due to strong progress in the development of Stirling-cycle cryocoolers, portable HPGe spectrometers with direct electromechanical cooling have been also created [4–6].
Both cooling methods mentioned above have their limitations associated with the possible unavailability of liquid nitrogen in the first case or with the increased weight of the instrument and possible degradation of the energy resolution of an HPGe detector due to microphonics induced by an electromechanical cryocooler in the latter case.
To overcome these difficulties a portable HPGe spectrometer with an alternate hybrid cooling system is proposed in this work.
One of the applications in which optimal spectrometric characteristics of the instrument along with the highest availability and portability of the device are of key relevance is a non-destructive verification of U enrichment and Pu isotopic composition by nuclear safeguards. To solve this task of analysis portable liquid nitrogen cooled and to a lesser extent, HPGe spectrometers with direct electromechanical cooling are currently used.
The instruments are accompanied by specialized isotopic analysis software codes which use intrinsic detection efficiency curves and reference nuclear data (gamma and X-ray energies and branching ratios) for U and Pu isotopic composition determination and do not require calibration standards [7, 8]. The most accurate results can be achieved using a low energy region of the spectrum, around 100-keV, and for the best performance in deconvolution of the peaks in this region, optimal energy resolution of HPGe detector is required. Therefore, a planar HPGe detector cooled by liquid nitrogen is currently a detector of choice for the given application.

The goal of the present research was to demonstrate a viable alternative to the traditional cooling methods of HPGe detectors by developing an apparatus able to simplify the verification activities of nuclear inspectors keeping the highest quality of the inspection results.

2. Historical overview

In the 1970th a borehole probe with Ge(Li) detector cooled by melting propane was developed [9]. The goal was to extend the usage of germanium detectors for the applications where boil-off of liquid nitrogen is precluded. In the borehole spectrometer, the propane was pre-cooled to the solid-state by liquid nitrogen. At ambient temperature and normal pressure, propane is gas but during cooling, it becomes first liquid and then solid. At this point, while propane melts the device can be used for the measurements.
The melting temperature of propane is 85 K what is low enough for Ge detector cooling, and while the propane melts the temperature remains stable what is essential for proper detector operation.
Although in the 1970th characteristics of HPGe detectors depending on operating temperature have been studied [10] and it was found that HPGe detectors could operate at temperatures as high as up to 150 K, what makes the application of cryogens with a much higher melting point compared to the melting temperature of propane possible.
Conceptual considerations on the use of other cryogens with a higher melting point were made in the Doctoral thesis by Earnshaw in 1987 and the properties and suitability of isopentane for HPGe detector cooling were studied and evaluated [11].
In the 1990th a deep-water gamma spectrometer cooled by melting propane was constructed [12], and an intermediate cooling substance between HPGe detector and liquid nitrogen such as propane was called a cryoaccumulator. This is a good name which reflects the fact that the cooling power is provided using the energy barrier accumulated at the phase transition from the solid to the liquid state.

3. Concept of the instrument

At the modern stage, due to the availability of compact electromechanical cryocoolers, it became possible to use a cryocooler instead of liquid nitrogen to cool down a cryogen to the solid state. As a result of the present research and development described in this paper, a hand-held HPGe spectrometer with a such hybrid cooling system is constructed.
The cooling system of the instrument contains a cryogen which is cooled down to a solid crystalline state by a detachable Stirling-cycle cryocooler, so after the cryogen cooling a cryocooler is disconnected from the main measuring device. Such cooling solution is liquid nitrogen-free and has advantages over direct electrical cooling in terms of the weight of the instrument (Figure 1) and spectrometric characteristics of the HPGe detector (no deterioration of energy resolution).

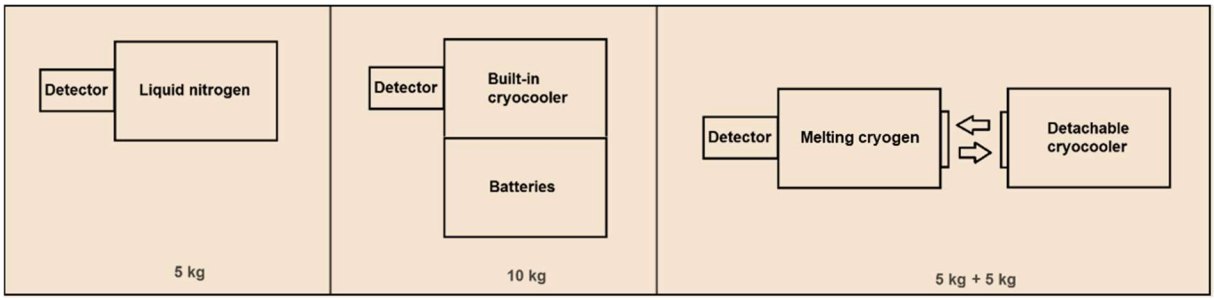

Figure 1 – Comparison of traditional cooling systems (left and middle) with the developed one (right).

The developed instrument (Figure 2) is a complete hand-held spectrometer based on an HPGe detector with an integrated preamplifier, an amplifier, an HV power supply, a multichannel analyzer and on-board rechargeable batteries for powering the spectrometric chain.
The instrument contains a planar HPGe detector and was created for the U and Pu isotopic composition verification by nuclear safeguards using low-energy X-ray region and MGA, MGAU and FRAM software codes. The instrument may also contain a coaxial HPGe detector providing higher detection efficiency at the energies over 100-keV, so as to be more suitable for the measurements of U in the enrichment meter mode (using an intensity of 185.7-keV peak) and Pu using high-energy regions (i.e. up to 1-MeV).

Figure 2 – Radionuclide Identifier for Gamma Analysis – RIGA.

4. Choice of melting cryogen

When the cooling system with melting cryogen is considered, then for the best performance it is essential to find the highest possible operating temperature of the HPGe detector. Also depending on the task of analysis it is important to know at which point the deterioration of energy resolution occurs, in which temperature range the energy resolution might be acceptable and at which point the energy resolution is totally unacceptable or the detector simply stops to operate due to the high leakage current and saturation of the spectrometry chain. In other words, the curve of the energy resolution vs. detector temperature shall be measured.

4.1. Energy resolution vs. detector temperature

To study the effect of temperature on the energy resolution of an HPGe detector, a planar HPGe detector was used. The detector had a miniature Dewar vessel with a volume of 600 ml [2]. Isopentane was first filled in the Dewar vessel and then it was transferred to the solid state by direct cooling with liquid nitrogen.
The detector temperature was measured using a Pt100 temperature sensor attached to the detector holder. In order to reduce heat flow through radiation and get HPGe detector temperature as much close as possible to the isopentane temperature, the detector in the holder was wrapped with a multilayer insulation made from aluminized mylar.

At the isopentane melting point of 113 K the energy resolution for Co-57 was the same as the energy resolution at 77 K and was equal to 570-eV. The curve shown in Figure 3 below was obtained during isopentane warm-up from melting to higher temperatures.

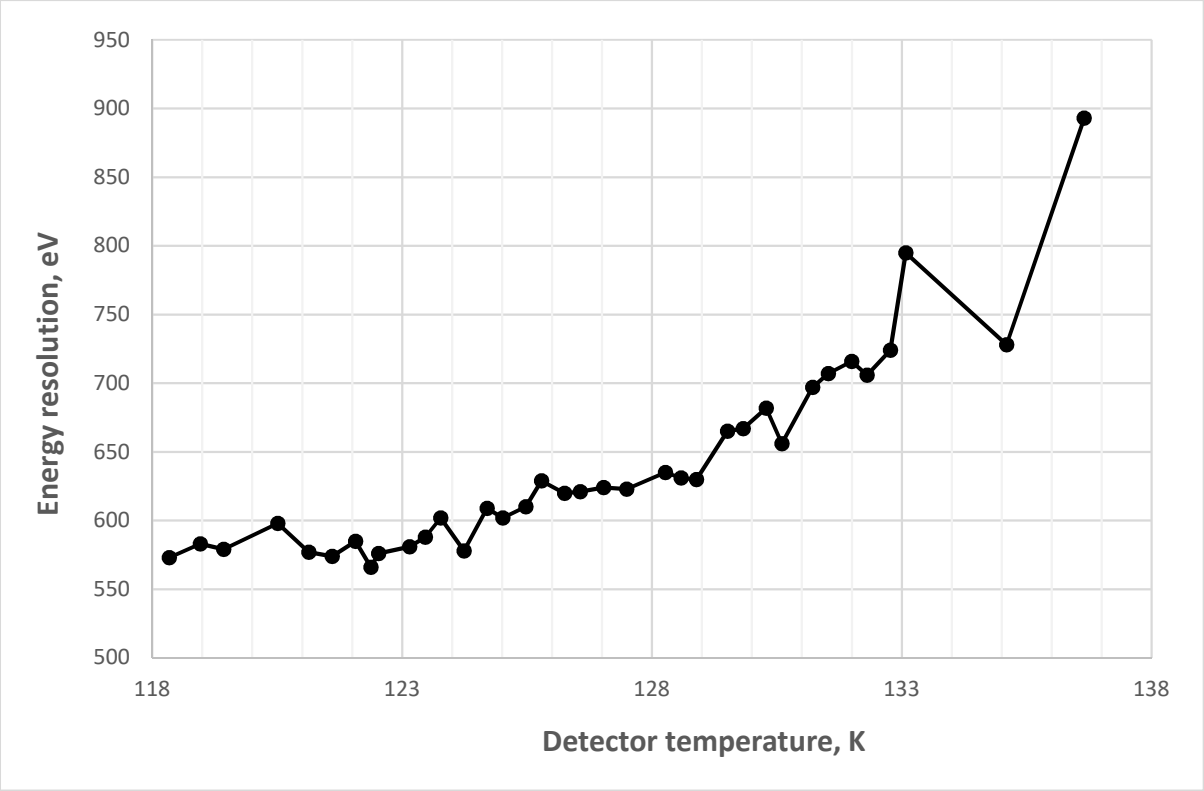

Figure 3 – Energy resolution at 122-keV (Co-57) vs. planar HPGe detector temperature.

4.2. Properties of the melting cryogen

The melting point of propane is 85 K and the corresponding enthalpy of fusion is 3.5 kJ/mol, for the comparison boiling point of nitrogen is 77 K and the corresponding enthalpy of vaporization is 2.79 kJ/mol. In terms of the holding time use of solid propane in the cooling system provides little advantage over the use of liquid nitrogen. But, the use of isopentane having a higher melting temperature of 113 K and higher enthalpy of fusion of 5.15 kJ/mol provides a real advantage for the holding time for the same mass of cryogen. In Table 1 are shown physical properties of the selected cryogens sorted by the melting temperature.

Table 1 – Physical properties of the selected cryogens.

#	Substance	Melting point, K	Enthalpy of fusion, kJ/mol	Boiling point, K	FWHM at 122 keV, eV
1.	Propane	85	3.5	230.8	570
2.	Isopentane	113.2	5.2	303	570
3.	Isoprene	127	4.9	307	620
4.	Methylcyclopentane	131	6.9	345	700
5.	1-Hexene	133.2	9.4	333	750

It is worth mentioning that the cryogen should have high purity to achieve the crystalline state. Although it is essential to have a high boiling point to have low vapor pressure at room temperature in a closed-type Dewar vessel and therefore shorten the requirements to the pressure vessel design, therefore only cryogens with high boiling points are listed in the Table. Corresponding values of the energy resolution for a given melting temperature are shown in the last column.
In the cooling system of RIGA, a closed-type Dewar vessel with encapsulated cryogen is implemented, and in order to achieve optimal energy resolution isopentane is used as a cryogen. However, depending on the task of the analysis, the cryogen in the cooling system can be easily substituted.

5. Cryostat

5.1. Basic characteristics

The industrial cryostat of the RIGA is shown in Figure 4. The outer vessel of the cryostat Dewar is made from steel, the inner vessel from oxygen-free copper. The volume of a closed-type Dewar vessel is 800 ml. The cryostat is designed with a single O-ring seal placed under the endcap flange. The endcap is made from aluminium. The electrical and HV feedthroughs are welded to the body of the cryostat.
For the U/Pu measurements, the detector should be collimated and the optimal method in terms of the reduction of the weight of the instrument is to use a high-Z collimator with appropriate X-ray liners placed directly around the detector holder inside the cryostat [1].
In this case, tungsten is a preferred material for the collimator also because of its low-outgassing vacuum properties.

Figure 4 – Cryostat of the RIGA.

5.2. Thermal characteristics

As the conductive heat flow defined by equation 1 depends on the difference between the hot and cold end of a thermal bridge between the inner vessel of the Dewar filled with the isopentane and the outer vessel, for the best holding time, the neck of the Dewar shall be made from the material of low thermal conductivity and shall have a small wall thickness. And it is desirable to select a cryogen with the highest possible melting point at which the energy resolution doesn't deteriorate.
The neck of the Dewar vessel of the cryostat is made from stainless steel with a wall thickness of 0.38 mm. Calculated heat flow through the neck is 1.25 W at 113 K and ambient temperature of 300 K. The choice of stainless steel for the neck over fiberglass was made deliberately, to achieve the lowest possible outgassing rate and therefore to provide the highest possible lifetime of the instrument without vacuum servicing.

$$Q = \frac{S}{h} \cdot \lambda \cdot (T_2 - T_1) \qquad (1)$$

Q – Heat flow through conduction, W
S – Normal cross section of a thermal bridge, m^2
h – Length of a thermal bridge, m
λ – Thermal conductivity of the material of the thermal bridge, W/(m·K)
$T_2 – T_1$ –Temperature difference, K

Heat flow through radiation is defined according to the equation 2. To decrease the heat flow through radiation a multilayer insulation was applied around the inner vessel filled with isopentane.

$$Q = 5{,}7 \cdot \varepsilon \cdot \left[\left(\frac{T_2}{100}\right)^4 - \left(\frac{T_1}{100}\right)^4\right] \cdot F \cdot E \qquad (2)$$

Q – Thermal flow, W
F – Surface of the inner cylinder, m^2
ε – Emissivity factor, $W / (m^2 \cdot K^4)$
T_2, T_1 – Temperatures of outer and inner cylinders, K
E = 1

It is estimated that the heat flow through radiation is less than 0.5 W.
The experimental value obtained for the cumulative heat flow (conduction plus radiation) using the melting time of a given mass of isopentane is 2.47 W at 113 K and ambient temperature of 300 K.
The difference between calculated and measured values can be attributed to the absence of a multilayer insulation around the neck and small spacing between a part of the neck and a wall of the outer vessel.

5.3. Vacuum characteristics

As the cryostat has low vacuum volume and a high surface-to-volume ratio, the pressure build-up due to outgassing can be very fast, so the accent was made on the provision of the cryostat with long vacuum life by utilizing low-outgassing materials. Another factor that was considered is the mechanical strength of the neck. Therefore, the neck was made from stainless steel. The vacuum support system of the instrument contains a sorption pump based on activated charcoal, hydrogen getters and ion pump. The measured ultimate pressure in the cryostat is 1.2×10^{-8} Torr. It is by three orders of magnitude better than the pressure required for the proper operation of a multilayer insulation, what gives a good reserve.
As a low-outgassing alternative, the neck can be made from a Hastelloy C-276; in this case, a two times improvement in thermal flow through conduction over stainless steel will be provided.
For the neck made from 0.8 mm thick fiberglass reinforced plastic, as traditionally made in the detector Dewar's, the heat flow through the neck will be reduced by one order of magnitude, and significant improvement in terms of the holding time will be achieved. A shortlist of suitable alternatives for the material of the neck is given in Table 2.

Table 2 – Thermal conductivity of selected materials.

Material	Thermal conductivity, W/(m·K)
Stainless steel (AISI 304)	16,2
Fiberglass G-10	0,4
Hastelloy C-276	8.1

6. Cooling system

The cooling cycle of the RIGA is illustrated by the temperature curve in Figure 5. The RIGA with a detachable docking station based on the Stirling-cycle cryocooler is shown in Figure 6. The cooling power of the cryocooler is 5 W at 77 K and 10 W at 113 K, at a heat rejection temperature of 300 K.

The measured curve has the following critical points and segments:

a) At point 1 the detector is warm; isopentane is in the liquid state; the cryocooler is just turned on;
b) At point 2 isopentane is already cooled to the solid state (cooling time = 7 hours). At this point, the RIGA can be detached from the docking station and the measurement can be done, or the cooling power of the cryocooler can be reduced to maintain the existing temperature;
c) Between points 2 and 3 isopentane is overcooled;
d) At point 3 the cryocooler is switched off;
e) Between points 3 and 4 isopentane is still overcooled but the temperature is rising;
f) At point 4 melting temperature of isopentane is reached;
g) Between points 4 and 5 isopentane is melting and the temperature is stable (holding time = 4 hours);
h) At point 5 and onwards the isopentane is warming up and is liquid.

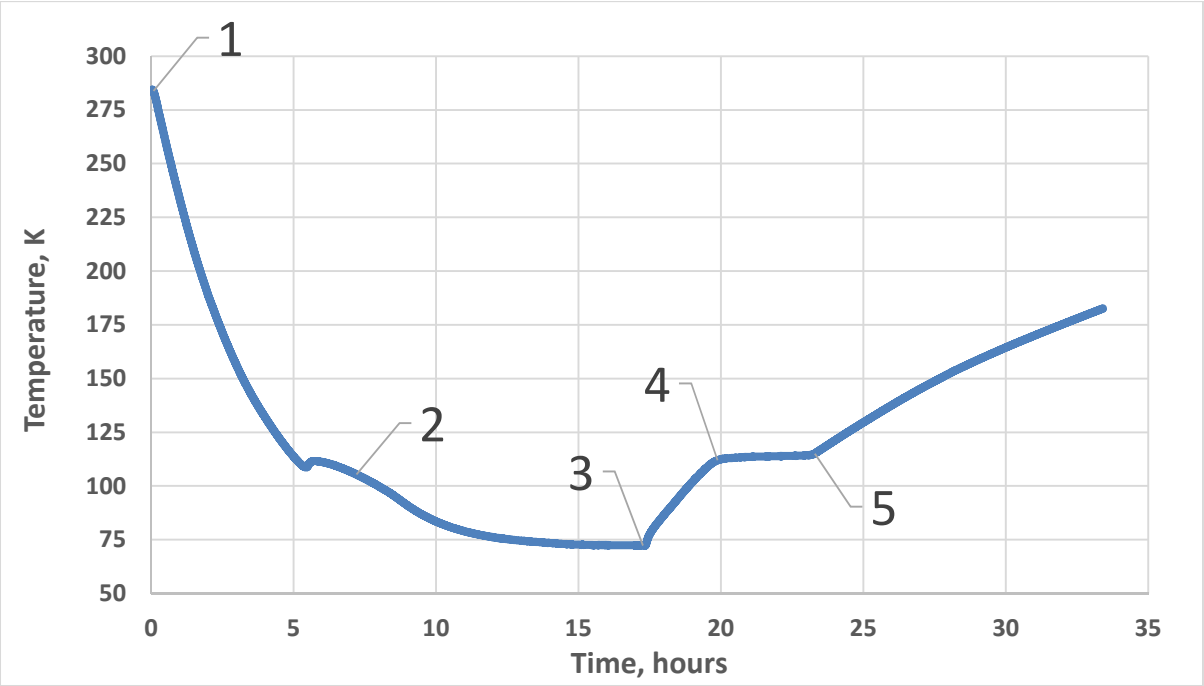

Figure 5 – Cooling cycle.

Figure 6 – RIGA with the docking station.

7. Spectrometer

The instrument is a complete hand-held HPGe spectrometer which contains a built-in preamplifier with HV inhibit circuit, an amplifier, a 16k channel MCA, an HV power supply – 4500V, and onboard batteries to power the spectrometric chain for 10 hours before re-charging. The multichannel analyzer is an OEM version of MCA-527 [13]. The electronics section is located below the cryostat Dewar in a housing made from anodized aluminium alloy which has a black colour. In Figure 7 the spectrometric boards of the MCA are taken out from the electronics housing for illustration.

Figure 7 – RIGA with the MCA shown outside of the electronics housing for illustration.

Summary of the technical characteristics of the instrument are shown in Table 3.

Table 3 – Main characteristics of the RIGA.

#	Parameter	Value
1.	Energy resolution of a planar HPGe detector	570 eV at 122 keV 480 eV at 59.5 keV 1230 eV at 662 keV
2.	Built-in spectrometry chain	Yes
3.	Cooling time of the cryogen	7 hours
4.	Autonomous operation time	4 hours
5.	Dimensions of the RIGA, D x W x H	364 × 92 × 239 mm
6.	Weight of the RIGA (without the docking station)	3.9 kg
7.	Weight of the docking station (with the power supply)	4.5 kg
8.	Instrument is powered by internal battery and controlled by Tablet PC	Yes

8. Conclusion

A hand-held HPGe spectrometer with a hybrid cooling system is developed.
A cooling system of the spectrometer is liquid nitrogen-free and provides optimal energy resolution for a planar HPGe detector. It contains a melting cryogen, which is pre-cooled to the solid crystalline state by a detachable Stirling-cycle cryocooler.
When cooled the cryogen works as a thermal buffer keeping the temperature of the detector low enough and stable using the energy barrier accumulated at the phase transition from the solid to the liquid state.
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The instrument is created for nuclear safeguards inspectors for such applications as U enrichment measurements and Pu isotopic composition verification with MGA, MGAU and FRAM software codes.

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