

**Saad Dahleb Blida University -1-**  
**Faculty of Sciences**  
**Physics Department**



# Memory

In order to obtain the diploma of

**MASTER IN PHYSICS**

**Speciality: APPLIED PHYSICS**

Title of the memory

**Application of Gamma spectrometry to radio-isotopic control of  
foods**

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## ملخص:

تستورد الجزائر جزءاً كبيراً من احتياجاتها الغذائية. لذلك لابد من مراقبة الغذاء المستورد من أجل حماية الصحة العامة، ثم الإعلان عن ملاءمته للاستهلاك البشري. يجب أن يمتثل الغذاء لقواعد معينة من بينها عدم وجود تلوث إشعاعي به. يقوم مركز البحث النووي بالجزائر بتحديد و تقدير النظائر المشعة المتواجدة في الاغذية الصادرة و المستوردة باستخدام تقنية قياس طيف غاما.

**الكلمات المفتاحية:** النشاط الإشعاعي- قياس طيف غاما- النظائر المشعة- تحليل النظائر المشعة- تلوث غذائي.

## Résumé:

L'Algérie est un pays qui importe une grande partie de ses besoins alimentaires. Dans un but de protection de la santé publique, les aliments importés doivent être contrôlés et déclarés propres à la consommation humaine. C'est pourquoi, ils doivent répondre à un certain nombre de règles dont l'absence de contamination radioactive.

Le Centre de Recherche Nucléaire d'Alger (CRNA) identifie et détermine les concentrations des radio-isotopes trouvés dans les aliments importés et exportés à l'aide de la spectrométrie gamma.

**Mots clés :** radioactivité – spectrométrie gamma – radio-isotopes – analyse des radio-isotopes – contamination alimentaire.

## Abstract:

Algeria imports a large part of its food needs. Imported food must be monitored to protect public health, and then matched to human consumption. Therefore it must fit with certain rules, including the absence of radioactive contamination.

The Research Nuclear Center in Algiers (CRNA) identifies and estimates radioactive isotopes found in imported and exported foods using gamma-ray spectrometry.

**Key words:** radioactivity – gamma spectrometry – radio-isotopes – radio-isotopes analysis – food contamination.

# Appreciation

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## **ABBREVIATIONS LIST**

**ICRP** International Commission on Radiological Protection

**IPF** import to product factor

**FAO** Food Alimentation Organization

**COMENA** COMmissariat à l'ENERgie Atomique « Atomic Energy Commission »

**CCFAC** Codex Committee on Food Additives and Contaminants

**CRNA** Centre de Recherche Nucléaire d'Alger «Nuclear Research Center of Algiers»

**WHO** World Health Organization

**ADC** Analog-to-Digital Converters

**MCA** Multi-Channel Analyzer

**FWHM** Full Width at Half Maximum

**MDA** Minimum Detectable Activity

**ROI** Region Of Interest

## **INTRODUCTION**

Consumer protection and safe food production are among the main tasks of the relevant authorities in every country. The food quality can be influenced by many factors and one of them is certainly the level of radioactivity.

Detailed knowledge of the activity concentrations of radionuclides in foodstuff is necessary to determine the food quality, high concentrations give evidence that there are potential risks to human health thus emergency steps are needed.

In the framework of this memory, we have implemented a nuclear application that goes under the name of Gamma-ray spectrometry. This non-destructive technique is the most common one for measuring radioactivity in environmental samples and foodstuffs.

Milk is a basic substance for general consumption. It is consumed frequently either row or as component in other foodstuff such as cookies, ice cream, yogurt, chocolate, powdered chocolate and many others human aliments. Thus, we chose it as our work's subject, we selected powder milk originating from 13 producing countries.

The objective of this work is to ensure the radioactive non-contamination of imported powder milk by performing qualitative and quantitative analysis by gamma spectrometry.

This memory is divided into six chapters:

In the first chapter, we present a review of the concepts of radioactivity and the different origins of ionizing radiation, and the citation of radiation protection standards for the monitoring of the radiological quality of foodstuffs.

The second chapter is dedicated to the process of radionuclide transfer in the food chain.

In the third one, we talk about the interaction between the gamma radiation and matter, and the physical phenomena resulted from that interaction.

As for the fourth chapter, we present the gamma-ray spectrometer with a hyper pure germanium detector.

The fifth chapter relates to the experimental work carried out at the level of the radio isotopic analysis service within the Nuclear Research Centre of Algiers (CRNA) where experimental measurements were carried out.

Finally, in chapter six we present, analyze and interpret the experimental results obtained.

We terminate this work with a conclusion that summarizes the methodology and results that were gained in the process of this work.

## Chapter I: CONCEPTS ON RADIOACTIVITY

Radioactivity is a natural phenomenon that has existed since the origin of the Universe when atoms were first formed. Not all atoms have the same properties: some are stable and remain indefinitely identical to them while others are unstable. In order to obtain a better stability, these so-called radionuclides expel at a given moment a quantity of energy, in the form of radiation and/or particles: this phenomenon is called «radioactivity».

### 1 History

The radiation was first discovered by the director of the Wurzburg Physics Institute, the Prussian professor Wilhelm Röntgen (1845-1923) on November 8, 1895. He noticed a surprising phenomenon; the screen placed nearby seemed to shine with a green light. Moreover, his hand placed behind the screen showed the shadow of his hand bones. At the end of December he published a short article claiming the existence of an unknown and strange radiation that was quickly named “X-rays.” For this discovery, he received the first Nobel Prize in physics in 1901.

Subsequently, the French scientist Antoine Henri Becquerel (1852-1908) discovered natural radioactivity, while studying the existence of a possible relation between those famous X-rays and the fluorescence phenomena. At that time, he was studying the fluorescence of uranium salts. At first, he assumed that the salt would radiate X-radiation-like after exposing it to sunlight and showing fluorescence. However, later, by coincidence he noted that the photographic plates were impressed with no fluorescent uranium. The shadow of a copper cross that Becquerel had placed between the uranium and the covered plates was visible. The new radiation had not gone through it; Becquerel called them “U-rays”.

And that was the start of a new revolution in science that made the 20th century completely different from the previous ones. Many scientists followed Roentgen and Becquerel’s path, such as Marie Sklodowska Curie (1867-1934) and her husband Pierre Curie (1859-1906), the two run together a number of researches that ended up with a number of significant discoveries in the nuclear world.(Pöschl and Nollet 2007)

### 2 Generalities

The atom is the smallest particle of a chemical element that can exist; an atom includes a nucleus and a series of orbital circles that surround this nucleus similar to the solar system structure. The protons (**p**) and the neutrons (**n**) reside in the nucleus, the electrons ( $e^-$ ) are in the orbitals. The amount of protons in the nucleus determines the atomic number (**Z**), while (**A**) is the mass number; where:

$$A=Z+N\text{.....(1.1)}$$



Figure I.1: The atomic symbol

### 2.1. Definition of radioactivity

Radioactivity refers to the particles or electromagnetic waves which are emitted from nuclei as a result of nuclear instability (Anne Marie 2019). The most common types of radiation (also known as ionizing radiation) are called Alpha, beta, neutron particles, gamma and X-rays:

- \* Alpha radiation( $\alpha$ ): it occurs when an atom undergoes radioactive decay (a spontaneous change within the nucleus of an atom which results in the emission of particles or electromagnetic radiation (Gordon 2008), giving off a particle (called an alpha particle) consisting of two protons and two neutrons (essentially the nucleus of a helium-4 atom), changing the originating atom to one of an element with an atomic number 2 less and atomic weight 4 less than it started with. Due to their charge and mass, alpha particles interact strongly with matter, and only travel a few centimeters in air.(Mirion 2015)
- \* Beta radiation ( $\beta$ ): it takes the form of either an electron ( $e^-$ ) or a positron ( $e^+$ ) being emitted from an atom. Due to the smaller mass, it is able to travel further in air, up to a few meters, and can be stopped by a thick piece of plastic, or even a stack of paper.(Mirion 2015)
- \* Gamma radiation ( $\gamma$ ): unlike alpha or beta, Gamma radiation does not consist of any particles, instead consisting of a photon of energy being emitted from an unstable nucleus. Having no mass or charge, gamma radiation can travel much farther through air than alpha or beta, losing (on average) half its energy for every 152,4 m. Gamma waves can be stopped by a thick or dense enough layer material, with high atomic number materials such as lead or depleted uranium being the most effective form of shielding. (Mirion 2015)
- \* X-rays: they are similar to gamma radiation, with the primary difference being that they originate from the electron cloud. This is generally caused by energy changes in an electron, such as moving from a higher energy level to a lower one, causing the excess energy to be released. X-Rays are longer-wavelength and (usually) lower energy than gamma radiation, as well.(Mirion 2015)
- \* Neutron radiation: it consists of a free neutron, usually emitted as a result of spontaneous or induced nuclear fission (subdivision of a heavy atomic nucleus into two fragments of roughly equal mass, the process is accompanied by the

release of a large amount of energy). Able to travel hundreds or even thousands of meters in air, they are however able to be effectively stopped if blocked by a hydrogen-rich material, such as water. Neutrons are, in fact, the only type of radiation that is able to turn other materials radioactive. (Mirion 2015)

It should not be surprising that there are many nuclear isotopes which are unstable and emit some kind of radiation. Isotopes of an element have nuclei with the same number of protons (atomic number) but different numbers of neutrons. Therefore, isotopes have different masses and different nuclear properties.

### 3. Dosimetric quantities and units

#### 3.1. Activity

The original unit for measuring the amount of radioactivity was the Curie (Ci)—first defined to correspond to one gram of radium-226:

$$1 \text{ Curie} = 3.7 \times 10^{10} \text{ radioactive decays per second} \dots\dots\dots (1.2)$$

In the International System of Units (SI) the Curie has been replaced by the Becquerel (Bq), where:

$$1 \text{ Becquerel} = 1 \text{ radioactive decay per second} = 2.703 \times 10^{-11} \text{ Ci} \dots\dots\dots (1.3)$$

#### 3.2. Radioactive period

Half-life, in radioactivity is the interval of time required for one-half of the atomic nuclei of a radioactive sample to decay, or, equivalently, the time interval required for the number of disintegrations per second of a radioactive material to decrease by one-half.

where:

$$T = \ln 2 / \lambda \dots\dots\dots (1.4)$$

$\lambda$ : Radioactive decay constant [ $s^{-1}$ ].

### 4. Radioactive Sources

#### 4.1. Natural sources

Apart from stable chemical elements, very low concentrations of radioactive elements occur naturally in the environment. We can divide these natural radionuclides into three categories according to their origin and formation: primordial radionuclides, secondary radionuclides, and cosmogenic radionuclides.

Primordial Radionuclides: they are the terrestrial heritage of original nuclear synthesis, those that we still deal with today are the survivors of this synthesis of the simple fact that their radioactive period is of the order of the age of the earth at least equal to  $4.5 \times 10^9$  years. The most widespread primordial radionuclide is K-40.

- \* **Secondary Radionuclides:** The decay of primary radionuclides (the one that the decay chain starts with) continuously gives rise to a number of secondary radionuclides. Natural radionuclides Th-232, U-238, and U-235 decay (by  $\alpha$  and later also  $\beta$  decay) into nuclei, which are also radioactive, much like their other decay products (i.e., radioactive decay chains). In nature, there are three radioactive decay chains: Th-232, U-238, and U-235. To a certain extent, these three natural decay chains are similar. They consist of isotopes of heavy elements mostly of  $\alpha$  radioactivity (a smaller part is also  $\beta$ ). Radon appears in the second half of the series; its decay products have a short half-life and disintegrate simultaneously by  $\alpha$  and  $\beta$  decay. Radon is a radioactive noble gas, one of the heaviest gases. All three natural decay chains result in stable isotopes of lead.
- \* **Cosmogenic Radionuclides:** They are natural radionuclides that currently originate by nuclear reactions when high-energy cosmic radiation passes through the Earth's atmosphere. Examples include radiocarbon (C-14) and tritium (H-3). (Pöschl and Nollet 2007)

### 4.2. Artificial sources

For the demands of research and technology, industry, and health services, the few radionuclides of natural origin are far from sufficient. Therefore we must produce radionuclides artificially. The artificial radionuclides can be produced by nuclear reactors, by particle accelerators, or by radionuclide generators. (Pöschl and Nollet 2007)

In this work, we will be dealing with the artificial type of radiation since the most important origin of contamination of foods is the nuclear accident or defects where radionuclides resulting from nuclear reactions happening inside the reactors in power plants accidentally release from reactor and reach spaces situated in the neighbor of the site.

## 5. Contamination and irradiation

### 5.1. Contamination

If radioactive material is not in a sealed source container, it might be spread onto other objects. Contamination occurs when material that contains radioactive atoms is deposited on materials, skin, clothing, or any place where it is not desired. It is important to remember that radiation does not spread or get "on" or "in" people; rather, it is radioactive contamination that can be spread. A person contaminated with radioactive material will receive radiation exposure until the source of radiation (the radioactive material) is removed.

- \* A person is externally contaminated if radioactive material is on the skin or clothing.
- \* A person is internally contaminated if radioactive material is breathed in, swallowed, or absorbed through wounds.

- \* The environment is contaminated if radioactive material is spread about or is unconfined.

## 5.2. Irradiation

Irradiation is when someone or an object get exposed to high-energy radiation from X-rays, gamma rays, neutrons, and other sources, unlike contamination; irradiated items does not have any direct contact with the radioactive materials, it all happens from distance. Thus, the greater is the distance between the source and the item, the lower is the radiation exposure.

## 6. Risks to human health from radioactive contamination

When radiation passes through matter, the component atoms may be ionized or excited. In the case of interaction with tissue, the ionization or excitation of molecules can pose a danger for livings cells and damage them. (Pöschl and Nollet 2007)

If radionuclides, particularly those that are artificially produced or originating in nuclear power engineering, are released into the environment through accident or some kind of defect in nuclear power plant, they can pose a real danger of radioactive contamination.

Human system is more likely to develop detrimental cancers and severe diseases after getting exposed to high doses of radiation or to low doses for long among of time, thus reducing exposure period, keeping distance from the radioactive sources and putting screens can minimize the potential risks. The radionuclides released from radioactive sources such as cesium and iodine are the main responsible of these detrimental diseases because of their high concentrations in area near relevant sources and their heavily use more than other radionuclides.(Pöschl and Nollet 2007)

## 7. Regulations

### 7.1. International regulations

- CODEX ALIMENTARIUS RECOMMENDED LIMITS FOR RADIONUCLIDE CONTAMINATION IN FOODS:

The guidelines were elaborated by the Codex Committee on Food Additives and Contaminants (CCFAC) in 1989, and later revised by the same committee in 2006 following a request of the International Atomic Energy Agency (IAEA)(CODEX 2011). The CCFAC stated the following:

- \* The guideline levels are established for a list of 20 radionuclides : hydrogen-3 (H-3); carbon-14 (C-14); technetium-99 (Tc-99); sulphur-35 (S-35); cobalt-60 (Co-60); strontium-89 and 90 (Sr-89 and Sr-90); ruthenium-103 and 106 (Ru-103 and Ru-106); iodine-129 and 131 (I-129 and I-131); uranium-235 (U-235); cesium-134 and 137 (Cs-134 and Cs-137); plutonium-238,239 and 240 (Pu-238, Pu-239, Pu-240); cerium-144 (Ce-144); iridium-192 (Ir-192); and americium-241 (Am-241). The 20 radionuclides were selected because of their importance into relation to uptake



in the food chain or because of quantities of these were contained in nuclear facilities or industrial radiation sources, which could potentially contaminate foods because of an accident or a malevolent act;

- \* The 20 selected radionuclides were divided in four groups according to their Dose Per Unit Intake i.e. contamination doses ingestion (DPUI);
- \* Naturally occurring radionuclides were excluded because the resource required calculating exposure would have been out of proportion to the health benefit achieved;
- \* An intervention exemption level of 1 mSv per year, in accordance with the International Commission on Radiological Protection (ICRP) recommendation;
- \* The assumption that 10% of the diet is of imported food all of which is contaminated, giving an Import to Product Factor IPF (the ratio of the amount of foodstuffs imported per year from areas contaminated with radionuclides to total amount produced and imported annually in the region of country under consideration) of 0.1, this was based on a statistical data of the Food Alimentation Organization (FAO) on production and import of all countries worldwide;
- \* Guideline levels may be increased by a factor of 10 for food consumed in small quantities (e.g. spices) and that represent only a small percentage of total diet.

The guideline levels are based on the IAEA most conservative values of the radionuclides-specific and age-specific.

The guideline levels are based on an assumed annual consumption of 500 kg for adults and 200 kg for infant. (CODEX 2011)

The Codex’s guideline levels for radionuclides in foods contaminated following a nuclear or radiological emergency for use in international trade are presented in the table I.1:

Product name	Representative radionuclides	Level in Bq/kg
Infant foods	Pu-238,239 and 240; Am-241	1
	Sr-90; Ru-106; I-131 and 129; U-235	100
	S-35;Co-60; Sr-89; Ru-103; Cs-134 and 137; Ce-144; Ir-192	1000
	H-3; C-14;Tc-99	
Foods other than infant foods	Pu-238,239 and 240; Am-241	10
	Sr-90; Ru-106; I-131 and 129; U-235	100
	S-35;Co-60; Sr-89; Ru-103; Cs-134 and 137; Ce-144; Ir-192	10000
	H-3; C-14;Tc-99	

Table I.1: The Codex’s guideline levels for radionuclides in foods (CODEX 2011)

## 7.2. Algerian regulations

- National texts:

Presidential Decree 05-117 relating to radiation protection measures ionizing (The Algerian republic, Official journal 2005), states in particular:

Art. 90. — The Atomic Energy Commission (COMENA) is responsible for the permanent control of radioactivity on the National territory.

They must ensure the assistance of the competent bodies for the establishment of the national radiation surveillance network. Under normal conditions, the planned control above includes:

- 1) Regular determination of air, water, soil and the food chain radioactivity.
- 2) Evaluation of the doses eventually received by the population.

The Atomic Energy Commission transmits the results and conclusions to the competent authorities.

Art. 92. — Foodstuff importers subject to prior checking on the levels of radioactive contamination have to go through this operation before the receipt of these products.

Art. 93. — Isotope analyzes are performed by the Atomic Energy Commission which is to be respond at the latest within forty eight hours (48h) on the levels of radioactive contamination.

Art. 94. — The Marketing and consumption of imported food products are subject to results indicating that contamination levels do not exceed the maximum tolerances fixed by decree of the minister in charge of trade.

Art. 95. —It is demanded from the Importers to require from their suppliers for each cargo a certificate of isotopic analyzes, delivered by the competent authority in the matter of the country of the goods, attesting to the level of radioactive contamination in the imported products.

Referring to the ministerial decree of 2 April 2000 amending and supplementing the decree of 27 October 1999 concerning the specifications of the industrial milk powder and the terms and conditions of its presentation, possession, use and marketing (The Algerian republic 2000), particularly in its seventh article, the maximum radioactive concentrations in the industrial milk powder are set the following:

Radionuclide	Radioactive activity (Bq/kg)
Americium-241	1
Plutonium – 239	1
Iodine – 131	67
Strontium – 90	67
Cesium – 134	202
Cesium – 137	267

Table I.2: Maximum radioactive concentration required by Algerian regulations for industrial milk powder

- ✓ The lower the value of the nuclides found in the milk powder, the better is the product's health value.
- ✓ When it comes to most food items, Algeria has adopted the international standards imposed by the various bodies responsible for monitoring radioactivity (IAEA, Codex Alimentarius and FAO).

## Chapter II: THEORETICAL PORTION

In our modern world, it is almost impossible to avoid getting in contact with radioactivity; artificial radionuclides always manage to find their ways to our daily life.

In this chapter, we will discuss Mechanisms for transferring radionuclides to food.

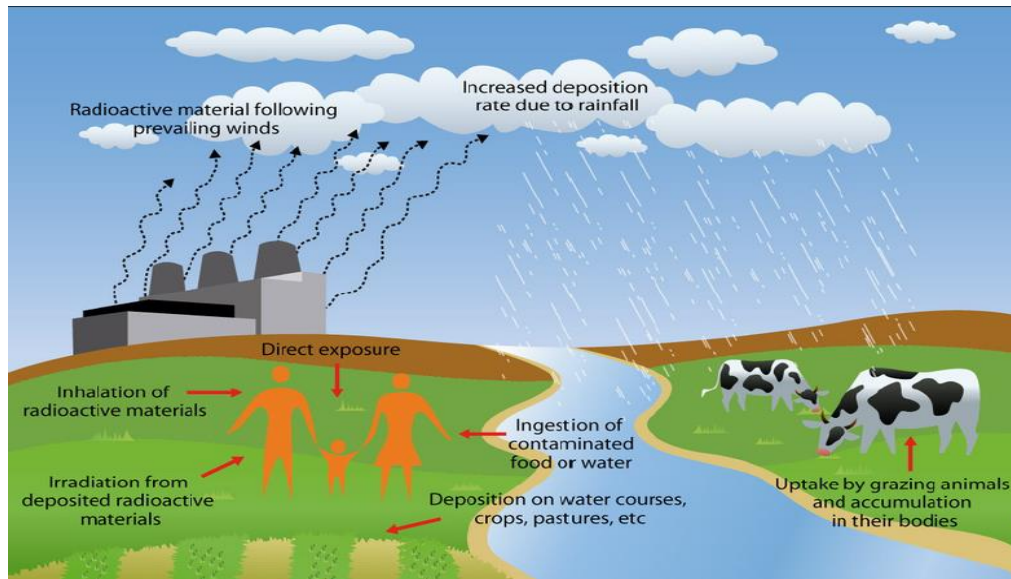


Figure II.1: Food chain contamination

### I. Radioactive contamination of the food chain

Background levels of radionuclides in foods vary and are dependent on several factors, including the type of food and the geographic region where the food has been produced. The common radionuclides in food are potassium-40 (K-40), radium-226 (Ra-226) and uranium 238 (U-238) and their associated progeny. In general, K-40 is the most commonly occurring natural radioisotope. In milk, for example, levels of K-40 measure around 50 Bq/L, and for meat, bananas and other potassium rich products, levels may measure at several hundred Bq/kg. Other natural radioisotopes exist in much lower concentrations, and originate from the decay of uranium and thorium.(WHO and FAO 2011)

When large amounts of radioisotopes are discharged into the environment, they can affect foods by either falling onto the surface of foods like fruits and vegetables or animal feed as deposits from the air or through contaminated rainwater/snow. Radioactivity in water can also accumulate in rivers and the sea, depositing on fish and seafood. Once in the environment, radioactive material can also become incorporated into food as it is taken up by plants, seafood or ingested by animals like (figure II.1) shows.

Although many different kinds of radionuclides can be discharged following a major nuclear emergency, some are very short-lived and others do not readily transfer into food. The radionuclides generated in nuclear installations and that could be significant for the food chain have been already included in chapter one. (WHO and FAO 2011)

Of immediate concern is iodine-131, it is distributed over a wide area, found in water and on crop land is rapidly transferred from contaminated feed into milk. However, iodine-131 has a relatively short half-life and will decay within a few weeks. In contrast, radioactive cesium which can also be detected early on, is longer-lived (Cs-134 has a half-life of about 2 years and Cs-137 has a half-life of about 30 years) and can remain in the environment for a long-time. Radioactive cesium is also relatively rapidly transferred from feed to milk. Uptake of cesium into food is also of long-term concern. Other radioisotopes that could be of long-term concern if released are strontium and plutonium. Strontium-90 has a half-life of about 29 years and plutonium has a much longer half-life than that (Pu-238: 88 years, Pu-239: 24100 years, Pu-240: 6564 years). However, both strontium and plutonium are relatively immobile in the environment and are of concern more locally, thus it is unlikely to cause a problem in international food trade in the immediate and medium-term.(WHO and FAO 2011)

Open-air vegetables and plants can be affected by the atmospheric release of radionuclides, resulting in radioactive contamination. Thus, radionuclides tend to be detected from leafy vegetables especially the ones with large leafy parts in the early phase after a nuclear accident. Milk is also associated with the early-phase contamination due to the rapid transfer of radioactive iodine and "relatively" rapid transfer of radioactive cesium from contaminated feed into milk.(WHO and FAO 2011)

Over time, radioactivity can also build up within food, as radionuclides are transferred through soil into crops or animals, or into rivers, lakes and the sea where fish and other seafood could take up the radionuclides. Foods collected from the wild, such as mushrooms, berries and game meat, may continue to be a radiological problem for a long time. (WHO and FAO 2011)

### **I.1. Health effects**

Consuming contaminated food will increase the amount of radioactivity inside a person and therefore increase their exposure to radiation, hence possibly increasing the health risks associated with radiation exposure. The exact health effects will depend on which radionuclides have been ingested and the amount being ingested.(WHO and FAO 2011)

**Radioactive iodine (I-131)** in food is of immediate concern due to its rapid transfer to milk from contaminated feed and its accumulation in the thyroid gland. I-131 has a relatively short half-life (8 days) and will therefore naturally decay over a short time frame. If radioactive iodine is breathed in or swallowed, it will concentrate in the thyroid gland and increase the risk of thyroid cancer. The uptake of radioactive iodine into the

thyroid gland can be decreased or prevented by ingestion of non-radioactive iodine, by taking potassium iodide pills. Once the thyroid is saturated with iodine, no further iodine can be incorporated. Iodized table salt should not be used as an alternative to potassium iodide pills as it does not contain sufficient iodine to saturate the thyroid, and high salt intake may have adverse health effects.

**Radioactive cesium (Cs-134 and Cs-137)**, in contrast to radioactive iodine, has a long half-life (Cs-134: 2 years, Cs-137: 30 years). Radioactive cesium can stay in the environment for many years and could continue to present a longer term problem for food, and food production, and a threat to human health. If cesium-137 enters the body, it is distributed fairly uniformly throughout the body's soft tissues, resulting in exposure of those tissues. Compared to some other radionuclides, cesium-137 remains in the body for a relatively short time. Like all radionuclides, exposure to radiation from cesium-137 results in an increased risk of cancer.

(WHO and FAO 2011)

Other radionuclides could be of concern, depending on the nature of the nuclear accident and release of specific isotopes.

**II. Interaction of  $\gamma$ -radiation with matter**

**II.1. Mechanisms of Interaction**

When Gamma rays interact with matter, these can be either absorbed completely or scattered either by an electron or a nucleus through number of processes. Many of these processes are quite infrequent and some have yet not been observed. In the energy domain 0.01 to 10 MeV, gamma rays interrelate with matter primarily through three processes namely; Photoelectric effect, Compton Effect and Pair production. The amount of particular type of interaction probably depends upon incident photon energy, scattering angle, nature of the target material and experimental conditions. (Gordon 2008)

**II.1.1. Photoelectric absorption**

Photoelectric absorption arises by interaction of the gamma-ray photon with one of the bound electrons in an atom. The electron is ejected from its shell (Figure II.2) with a kinetic energy,  $E_e$ , given by:

$$E_e = E_\gamma - E_b \dots \dots \dots (II.1)$$

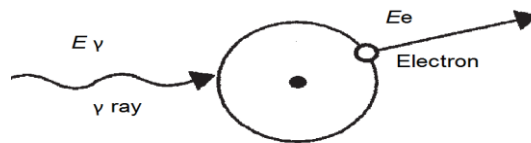


Figure II.2: The mechanism of photoelectric absorption

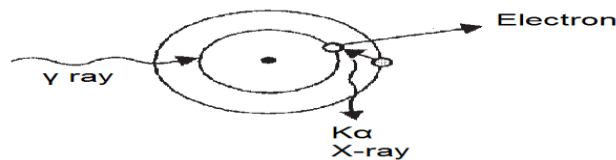


Figure II.3: Emission of fluorescent X-rays

where:

$E_\gamma$  are the gamma-ray energy and  $E_b$  the energy binding the electron in its shell. The atom is left in an excited state with an excess energy of  $E_b$  and recovers its equilibrium in one of two ways. The atom may de-excite by redistribution of the excitation energy between the remaining electrons in the atom. This can result in the release of further electrons from the atom (an Auger cascade). Alternatively, the vacancy left by the ejection of the photoelectron may be filled by a higher-energy electron falling into it with the emission of a characteristic X-ray which is called X-ray fluorescence (Figure II.2). This X-ray may then in turn undergo photoelectric absorption, perhaps emitting further X-rays which are absorbed, in turn, until ultimately all of the energy of the gamma-ray is absorbed. In order to conserve momentum when an electron is ejected, a very small amount of energy must be retained by the recoiling atom. This is very small and can be ignored for all practical purposes. The energy level from which the electron is ejected depends upon the energy of the gamma-ray. The most likely to be ejected is a K electron. If sufficient energy is not available to eject a K electron, then L or M electrons will be ejected instead. (Gordon 2008)

**II.1.2. Compton scattering**

Compton scattering (Figure II.4) is a direct interaction of the gamma-ray with an electron, transferring part of the gamma-ray energy. The energy imparted to the recoil electron is given by the following equation:

$$E_e = E_\gamma - E'_\gamma \dots\dots\dots(II.2)$$

Or:

$$E_e = E_\gamma - \left\{ 1 - \frac{1}{[1 + (1 - \cos \theta) E_\gamma / m_0 c^2]} \right\} \dots\dots\dots(II.3)$$

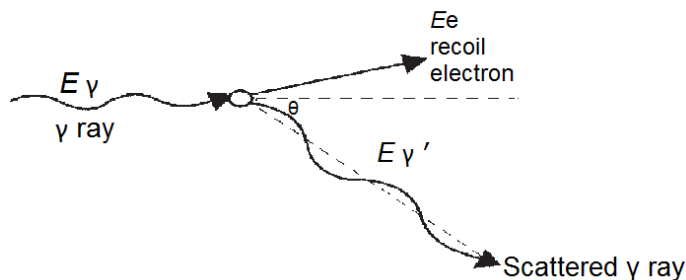


Figure II.4: The mechanism of Compton scattering

Putting different values of  $\theta$  into this equation shows how the energy absorbed varies with the scattering angle. Thus, with  $\theta = 0$ , i.e. scattering directly forward from the interaction point,  $E_e$  is found to be 0 and no energy is transferred to the detector. At the other extreme when the gamma-ray is backscattered and  $\theta = 180^\circ$ , the term within



brackets in the equation above is still less than 1 and so only a proportion of the gamma-ray energy will be transferred to the recoil electron. At intermediate scattering angles, the amount of energy transferred to the electron must be between those two extremes. (Gordon 2008)

### II.1.3. Pair production

Unlike photoelectric absorption and Compton scattering, pair production results from the interaction of the gamma ray with the atom as a whole. The process takes place within the Coulomb field of the nucleus, resulting in the conversion of a gamma-ray into an electron–positron pair. In a puff of quantum mechanical smoke, the gamma-ray disappears and an electron–positron pair appears. For this miracle to take place at all, the gamma-ray must carry energy at least equivalent to the combined rest mass of the two particles – 511 keV each, making 1022 keV in all. In practice, evidence of pair production is only seen within a gamma-ray spectrum when the energy is rather more than 1022 keV. In principle, pair production can also occur under the influence of the field of an electron but the probability is much lower and the energy threshold is 4 electron rest masses, making it negligible as a consideration in normal 0 to 3MeV gamma spectrometry. The electron and positron created share the excess gamma-ray energy i.e. the energy in excess of the combined electron–positron rest mass equally, losing it to the detector medium as they are slowed down. When the energy of the positron is reduced to near thermal energies, it must inevitably meet an electron and the two will annihilate, releasing two 511 keV annihilation photons. This is likely to happen within 1 ns of creation of the pair. The complete sequence of events is described in (Figure II.5). (Gordon 2008)

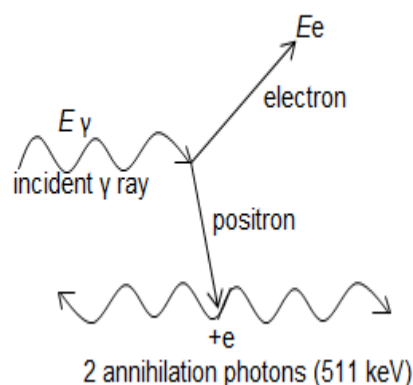


Figure II.5: The mechanism of pair production

## III. GAMMA-RAY SPECTROMETRY

### III.1. The gamma-ray spectrometry: Principle

The main purpose of gamma-ray spectrometry is to identify the different radionuclides constituting present in a radioactive sample and to quantify the activity of each radionuclide.

The principle of the detection of a gamma photon is based on the measurement of the energy that it has given up (partially or totally) to a sensitive material i.e. detector via one of the ionization mechanisms mentioned previously per unit of time.

The spectrum obtained is composed of peaks which constitute a graphical representation of the rate of pulses detected as a function of energy.(Gordon 2008)

### III.2. Detector

The interaction between the gamma radiation and the detector volume generates electrical charges which are collected thanks to the potential difference applied to the detector. There are two detector types in practice that are able to detect gamma rays over a wide energy range up to 12 MeV; these are the scintillation and the semiconductor detectors. To select between these two detector types one needs to consider several points. Industrial applications require durable, shock and vibration resistant, easy-to-use equipment, which may be operated in a wide range of temperature, humidity and pressure environments. These criteria can be better satisfied by scintillation detectors, and such detectors may also be satisfactory for monitoring the composition of nearly identical objects. The superior energy resolution of the semiconductor detectors makes them ideal for high sensitivity analysis of samples having a large variety of compositions. In this work, we will be focusing on the semiconductor detectors.(Tamas et Zsolt 2004)

#### III.2.1. Semiconductor Detectors

##### The Semiconductor:

In a free atom, the electrons are disposed in precisely determined energy levels. Combining a collection of atoms together into a solid structure broadens those energy levels into energy bands, each of which can contain a fixed number of electrons. Between these bands are energy regions that are forbidden to electrons. The upper most occupied energy band, the inhabitants of which are responsible for chemical reactions, is known as the valence band. In order for an electron to migrate within the material, it must be able to move out of its current energy state into another in order to move from atom to atom. If electrons can jump into suitable energy levels, then an external electric field applied to the material would cause a current to flow. There are three types of material: insulators, conductors and semiconductors. These differ in their electronic structures. The valence bands in the semiconductors are full but the band gap is much smaller, of the order of 1

eV. Under normal conditions there will always be a small population of electrons in the conduction band and the material will exhibit a limited degree of conductivity. The probability that an electron will be promoted to the conduction band is strongly influenced by temperature. (Gordon 2008)

$$P(T) \propto T^{3/2} \exp(-E_g/2Kt) \dots \dots \dots (II.1)$$

where:

T: temperature, in (°k)

K: Boltzmann constant

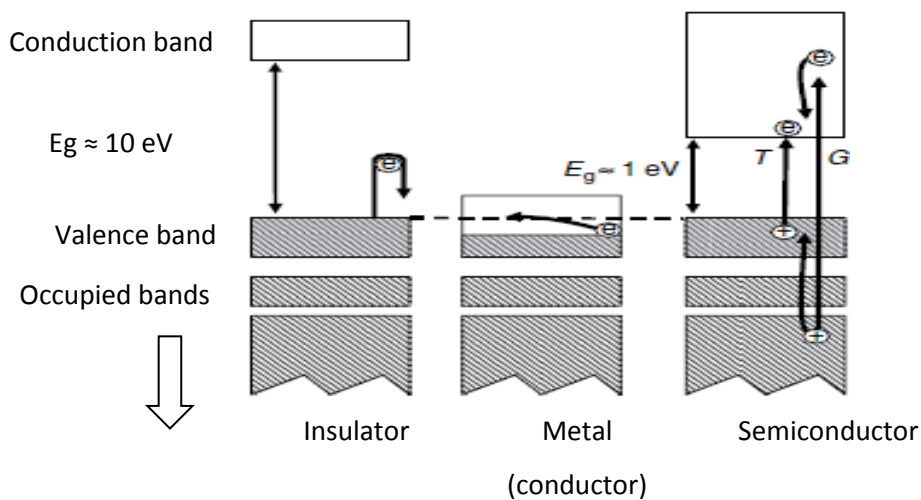


Figure II.6: Schematics of the electronic band structures in insulators, metals and semiconductors

**electron-hole pairs:**

When an electron is promoted from the valence due to some external energy in the form of heat energy, or any other, band to the conduction band, a vacancy is left behind in the otherwise full band. This vacancy is effectively positively charged and is referred to as a hole. Holes are also mobile. An electron within the valence band may replace that lost from the vacancy, thus filling the hole. That will leave, in turn, another vacancy. In the presence of an external electric field, the hole can appear to move towards the cathode (Figure II.7). Since both electrons and holes carry charge, both will contribute to the conductivity of the material. (Gordon 2008)

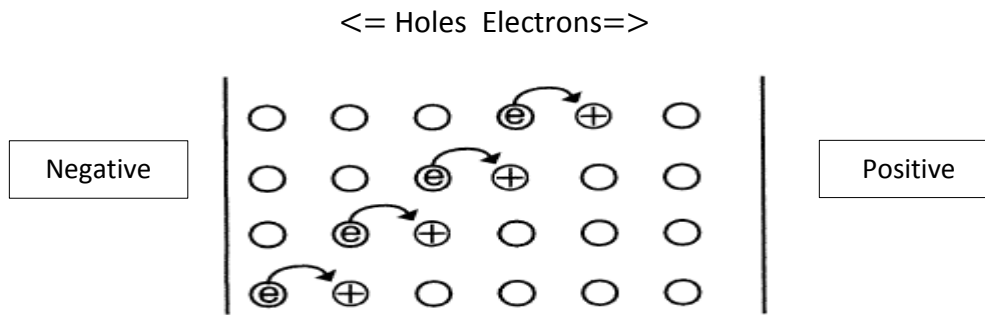


Figure II.7: A model for hole mobility in solid

### Operating:

The manner in which gamma radiation interacts with matter was explained in terms of various mechanisms in the previous chapter, each of which transfers energy from the gamma-ray to electrons, and in the case of pair production, positrons; these particles lose their kinetic energy by scattering around within the detector, creating ionized atoms and ion pairs. This population of secondary entities forms the basis of the detector signal. Having absorbed the gamma-ray and created many charged species (electron-hole pairs), the detector material must allow the charge to be collected in some manner and presented as an electrical signal. The most obvious way to do this is to supply an electric field across the detector material to 'sweep' the charge carriers out of the detector. This, of course, can only be done if the detector has suitable electrical characteristic. (Gordon 2008)

With that being said, we can summarize the operation of semiconductor detectors in the following points:

- \* Ionizing radiation enters the sensitive volume of the detector and interacts with the semiconductor material.
- \* Particle passing through the detector ionizes the atoms of semiconductor, producing the electron-hole pairs. The number of electron-hole pairs is proportional to the energy of the radiation to the semiconductor. As a result, a number of electrons are transferred from the valence band to the conduction band, and an equal number of holes are created in the valence band.
- \* Under the influence of an electric field, electrons and holes travel to the electrodes, where they result in a pulse that can be measured in an outer circuit,
- \* This pulse carries information about the energy of the original incident radiation. The number of such pulses per unit time also gives information about the intensity of the radiation.

### III.2.2. The germanium detector

The semiconductor gamma-ray detectors are made from either high purity germanium **Ge(HP)** crystals or lithium **Ge(Li)**. Lithium-compensated germanium detectors are manufactured in the same way as silicon compensated lithium detectors. This kind of detectors was marketed from the early 1960s until the early 1980s.(Fabien 2007)

It has been replaced by the hyper pure germanium **Ge (HP)** detectors whose performances are equivalent for the same active volume which allows in particular to be brought back to room temperature between measurements. In fact, since lithium diffuses at room temperature, Lithium-compensated germanium detectors cannot be used at room temperature. It's about a pure germanium crystal whose impurity concentration has been reduced from  $10^{13}$  to  $10^{10}$  atoms/  $cm^3$ . Germanium detectors exist in three geometric configurations: planar, coaxial and well (figure IV.3). The planar geometry allows for low detector response energy (from 1 keV to 1 MeV). It is with this configuration that the resolution is best (table II.1). The thickness of the active volume can be up to 1 or 2 cm, which gives a volume of 10 to 30  $cm^3$ , which is relatively low. (Fabien 2007)

The coaxial geometry (figure IV.3) can work around this problem; the active volume can reach 750  $cm^3$ . On the other hand, the energy response is shifted to the high energy: 10 MeV. In addition, the resolution is less good than detectors with planar geometry (table II.1). (Fabien 2007)

With the well geometry the detector response is in the same energy interval as coaxial geometry but the fact that a sample can be introduced inside the detector, this improves the detection.

The low energy resolution is less good than with coaxial, in the other hand in high energy geometry situation the resolution is better with well geometry. The germanium detectors are the ones that have the best performances for photon detection in a wide range of energy (20 keV to 1.5 MeV).(Fabien 2007)

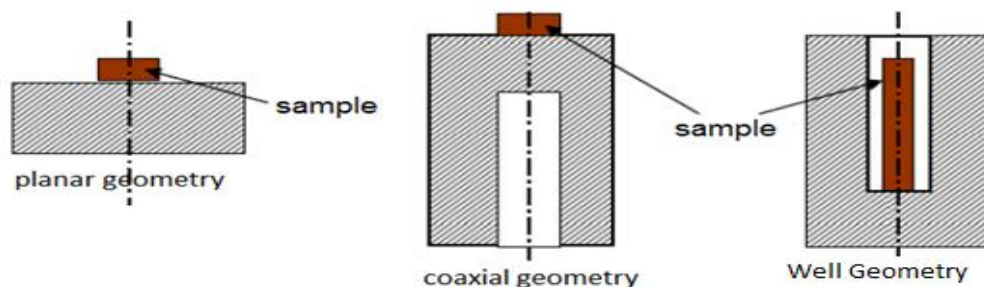


Figure II.8: Different geometric configurations of germanium detectors

Energy (keV)	Geometry		
	Planar	Coaxial	Well
122	0.65	0.80	1.20
1332	2.00	3.40	2.00

Table II.1: Resolution of a germanium detector in the three possible configurations

The germanium crystal has to be cooled to liquid nitrogen temperature (77 °K) when operated in order to minimize the leakage current that could destroy the detector. Also, at this temperature, the thermal noise is sufficiently low to assure good resolution.

**III.2.3. Main characteristics of a detector**

The quality of a gamma detector is related to its ability to detect rays even the small intense ones sometimes (efficiency criterion), and also to distinguish them together (resolution criterion). The following presents the different criteria that are generally used to characterize and compare the performance of the detector.

**III.2.4. The detection efficiency**

In general, the geometry of the detectors makes it impossible to measure all the radiation emitted. On the one hand, detectors rarely cover a sufficient solid angle, and on the other hand, some of the radiation passes through the detector without interacting or partially attenuates before leaving the useful detection area. The concept of detection efficiency has therefore been introduced which reflects the ability of a device to detect a given radiation of a given energy (Wassila 2011). Two types of efficiencies are defined as follows:

- The absolute efficiency or efficiency of a detector is the ratio of the measured radiation to the radiation emitted by the source. Efficiency is often expressed as a percentage of the number of radiation incidents. For particle counters, we have:

$$\epsilon_{abc} = \frac{\text{number of pulses counted}}{\text{number of radiations from the source}} \dots\dots\dots (II.2)$$

- Intrinsic efficiency only takes into account the radiation that has passed through the detector:

$$\epsilon_{int} = \frac{\text{number of pulses counted}}{\text{number of radiation received by the detector}} \dots\dots\dots (II.3)$$

**III.2.5. Dead time**

Electronics may introduce limitations on count rate due to dead time effects and load collection issues. Dead time is the minimum time to separate two events that will be transformed into two separate electrical signals. If a particle interacts with the crystal while the signal from a previous interaction is being acquired, it does not result in any counting because the input is blocked electronically. Dead time is linked to a limitation of the electronics: operations such as the conversion of the collected charge to the electrodes into an electrical signal, the amplification and filtering of this signal require a

fairly long time during which the signal is processed, knowing that only one signal. (Fabien 2007)

### III.2.6. Background noise

Background noise is the counting or recording of a spectrum under normal conditions of use of the measuring chain in the absence of the source whose radiation to be measured is also defined as the spectrum recorded from a detector, alone or with an empty container, the knowledge of this background noise is useful to improve the accuracy of the measurement results of the activity especially in the case of traces. The components of background noise are both origin cosmic radiation, radioactivity of radionuclides present in terrestrial materials (families of U-235, U-238 and Th-232) and artificial radioactivity that is present in the materials making up the spectrometer or in its immediate environment. (Wassila 2011)

### III.3. Presentation of a gamma-ray spectrometer chain

A gamma-ray spectrometer consists of the following elements:

- High voltage power supply
- A detector connected to a preamplifier.
- A preamplifier
- An amplifier.
- Analog-to-Digital Converters (ADC).
- Multi-Channel Analyzer (MCA).
- Computer.

The figure below represents the synoptic diagram of a gamma-ray spectrometer.

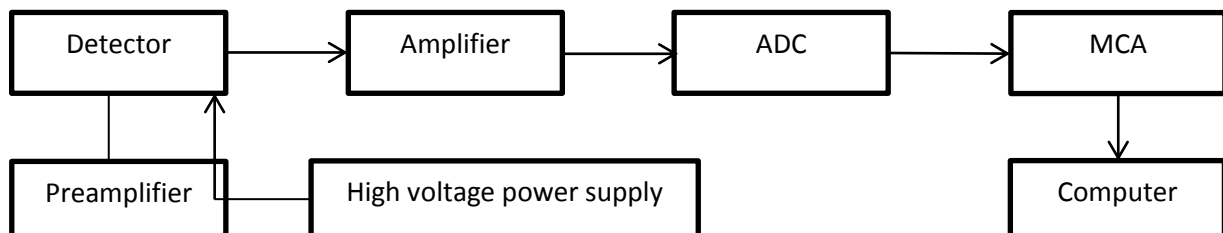


Figure II.9: The synoptic diagram of a gamma-ray spectrometer

#### III.3.1. High voltage power supply

The high voltage power supply is an essential element; it must fulfill the following conditions: first, be adjustable for the voltages imposed by the detector, in addition of having a very low noise and be able to withstand the current delivered by the voltage without voltage drop, the high voltage power supply must be adjustable for the voltages imposed by the detector, and also be stabilized so to not exhibit any drift over time. The purpose of the high voltage is to accelerate the electrons produced by the interaction of photons with the detector material.

### III.3.2. Preamplifier

The charge created within the detector by interaction with the gamma radiation is collected by the preamplifier. In spite of its name, the function of the preamplifier is not to amplify the pulse, it merely goes before the amplifier, but to interface the detector to the amplifier and collect the charge generated by absorption of the gamma-ray. It provides a high impedance load for the detector and a low impedance source for the amplifier. Preamplifiers, in general, can have various modes of operation: current-sensitive, voltage-sensitive and charge sensitive. Only the latter type is used in high-resolution gamma spectrometry using semiconductor detectors. It has advantages in terms of noise performance and because the gain is independent of detector capacitance.

### III.3.3. Amplifier

It uses the pulses coming from the preamplifier. First, it adapts the amplitude of the incident signal to a voltage range that can be used by the next stage. Then, it performs a shaping, most often Gaussian.

### III.3.4. Analogue-to-Digital Converters (ADC)

The ADC is used to convert the peak value of a pulse to a value digital. This value is the address at which the acquisition electronics will increment the content of one of an analyzer's channel

### III.3.5. Multi-Channel Analyzer (MCA)

The MCA allows sorting the impulses according to their amplitude. It has memories to store this data in channels corresponding to voltage intervals and transmit them online to a computer. The resulting records are histograms called "differentiated height spectra" most of the time, the software used for the acquisition data also allows for a first qualitative analysis and quantitative spectra.

### III.3.6. Signal processing and analysis software

Once transmitted to the main memory of the computer, the signals are processed by specialized software Génie 2000. The computer is designed to visualize the distribution of pulses, depending on their energies or the number of the corresponding channel, in the form of a spectrum. Each photon has a perfectly defined energy, so the energy spectrum is a line spectrum. The analysis of the spectrum can be divided into four main steps: the operations preceding the analysis of the spectrum; energy, efficiency and resolution calibrations of the measurement chain and the acquisition of a background noise spectrum. Then, the search for peaks followed by the corresponding analyzes and finally the identification of radionuclides and activity calculations. The software allows you to perform the most common functions. This is to define the acquisition time, we can set the real time or the actual time (live time), or else fix the total number of pulses and choose regions of interest (ROI). This function allows a region to be fixed which delimits the photoelectric peaks of interest formed by the interaction of photons with the



detector. It also allows us to calculate the area of the peak which allows us to go back to the concentration of the radionuclide.

## Chapter III: EXPERIMENTAL PORTION

As a major importer of powdered milk, we have chosen to analyze samples of powdered milk imported from thirteen different producing countries.

In this chapter, we will determine the concentration of the radionuclides of interest in the relevant samples using gamma-ray spectrometer.

### 1. The radio isotopic analysis service organization chart

The samples are received at the level of the radio isotopic analysis service at the Nuclear Research Center of Algiers (CRNA). Customers deliver their goods that are either imported from different foreign countries or meant to be exported. The goods go through many persuaders before they get analyzed using gamma-ray spectrometer, from receiving them to putting them in mini sacs to finally putting them into containers.

Once the analysis is over, a certificate proving the product is free of any radio-contamination is filled.

This last is provided to the customer in order to give him the permission to trade his product.

الجمهورية الجزائرية الديمقراطية الشعبية  
محافظة الطاقة النووية

Centre de Recherche Nucléaire d'Algier  
02, Boulevard Frantz Fanon, BP 399, Alger-gare, Alger  
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تيل: (213) 021 43 44 44 فاكس: (213) 021 43 42 80

N/Réf.: 1792/SA/DG/CRNA/2020 Alger, le \_\_\_\_\_

**CERTIFICAT  
DE NON-CONTAMINATION RADIOACTIVE**

La Directrice Générale du Centre de Recherche Nucléaire d'Algier certifie que l'analyse de l'échantillon, dont les spécifications sont données ci-dessous, effectuée par spectrométrie gamma ne révèle pas de contamination radioactive.

Echantillon(s) déposé(s) : Le \_\_\_\_\_ Par: \_\_\_\_\_  
 Nombre \_\_\_\_\_  
 Code interne \_\_\_\_\_ du \_\_\_\_\_  
 Demandeur \_\_\_\_\_  
 Référence de la demande \_\_\_\_\_ du \_\_\_\_\_  
 Importateur \_\_\_\_\_  
 Produits : Lait en poudre entier  
 Codes \_\_\_\_\_  
 Lot(s): \_\_\_\_\_ DF: \_\_\_\_\_ DP: \_\_\_\_\_  
 Origine \_\_\_\_\_  
 Navire \_\_\_\_\_ Du \_\_\_\_\_  
 Destination \_\_\_\_\_

Le présent certificat est délivré pour servir et faire valoir ce que de droit.

**La Directrice Générale**

Figure III.1: The certificate provided to customers

## 2. The experimental set-up

In this analysis, we used a CAMBERRA gamma spectrometry chain consisting of the following elements:

- A Ge(HP) type semiconductor detector mounted on a vertical cryostat maintained at the temperature of liquid nitrogen, with a the following physical characteristics:
  - Coaxial geometry open end, closed end facing window;
  - Diameter of 61mm;
  - Length of 46.5;
  - Distance from window (outside) of 6mm.
- High voltage power supply,
- A preamplifier,
- An amplifier,
- Integrated Analog-to-Digital Converter (ADC) with Multi-Channel Analyzer (MCA),
- A computer with data processing software (Genie2000),
- Calibration radioactive sources kit containing three radioactive sources (Am-241, Cs-137 and Co-60).



Figure III.2: Ge(HP) type semiconductor detector with coaxial geometry mounted on a vertical cryostat maintained at the temperature of liquid nitrogen

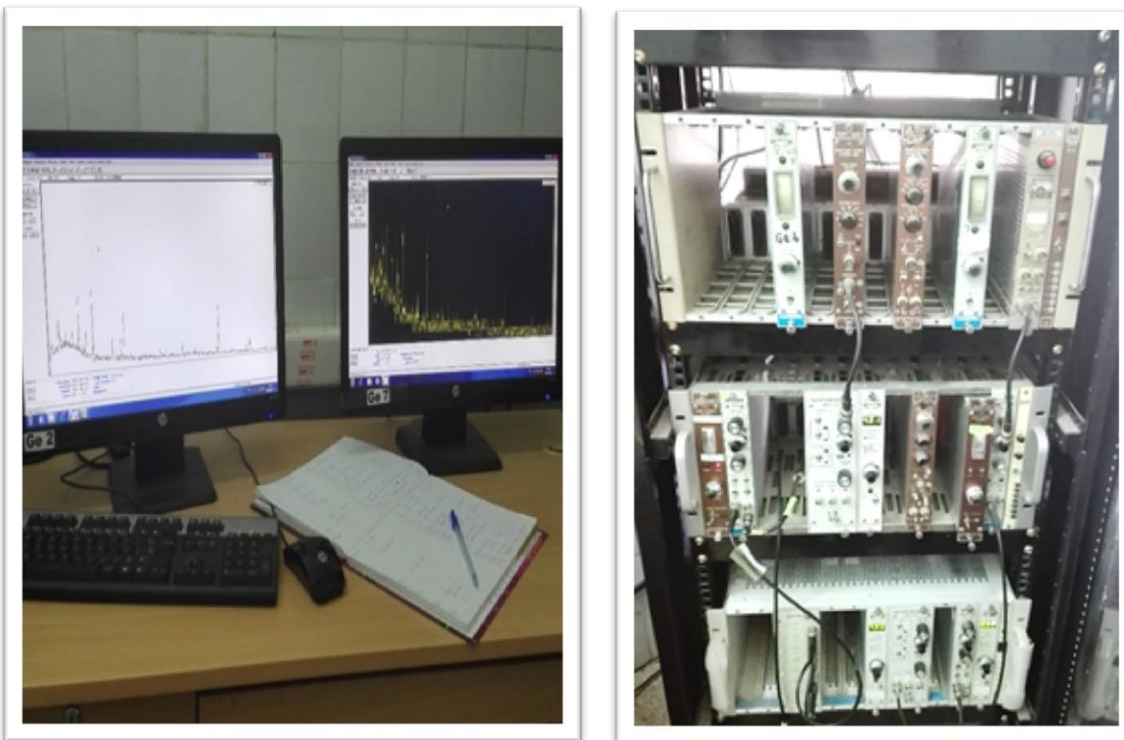


Figure III.3: Associated electronics

### 3. Gamma Spectrometry Chain Calibration

The calibration of the gamma spectrometry chain consists of two steps, the energy calibration which consists in verifying the linearity of the detection system to allow identifying the existing radionuclides in the sample to be analyzed, and efficiency calibration, which allows us to trace the activity of these radionuclides.

#### 3.1. Setting the Zero Pole

Before any study, it is necessary to make an adjustment of the Zero Pole for the corrections to zero the signal generated by the amplifier. This adjustment is made using an oscilloscope connected to the terminal of the amplifier to the MCA, to better join the return of the baseline. It allows having good Gaussian photoelectric peaks.

A deranged zero pole gives a Gaussian Under-shoot (twisted Gaussian to low energy) or Over-shoot (twisted Gaussian to high energy).

#### 3.2. Energy calibration

Before performing an energy calibration, the detector adjustment must first be performed by adjusting the amplifier gain taking into account the size of the MCA and ADC memory card, this step is called “conversion slope adjustment (keV/channel)”.

For this, we place a known mono-energy source (Cs-137, E= 661.7keV) in the detector for a 600 s acquisition time. Then, we adjust the top of the photoelectric peak to channel 3614 for a 16k (16384 channels) memory card with a maximum energy of 3000 keV. Thus, the conversion slope setting is 0.18 keV/channel.

For energy calibration, we used punctual gamma sources (Am-241, Cs-137, and Co-60) for an acquisition time of 7200 s; all the characteristics of these sources and the calibration results are illustrated in Table III.1. The choice of calibration sources is dictated by the spectral extent of the samples to be measured. Thus, Am-241 is used for low energies, Cs-137 is used for medium energies and Co-60 used for high energies. The three sources cover a total energy range of 3Mev.

The relationship between the channel numbers of the analyzer and the known energies of the emitted photons is carried out by suitable software (Genie 2000).

Source	Channels number	Energy (keV)	Intensity (%)	T <sub>1/2</sub> (year)
Am-241	325	59.5	35.90	432.20
Cs-137	3614	661.7	85.20	30.174
Co-60	6407	1173.2	99.90	5.271
	7277	1332.5	100.00	

Table III.1: Energy calibration results

Using the data from the standard sources, the resulting calibration line is given by the equation:

$$E = aC + b \dots \dots \dots (III.1)$$

where:

E: radionuclide energy expressed in keV;

C: channel number;

a: Slope of the line expressed in keV/channel;

b: Value of the origin's energy expressed in keV.

The energy calibration line of the measuring chain is given by Figure III.4.

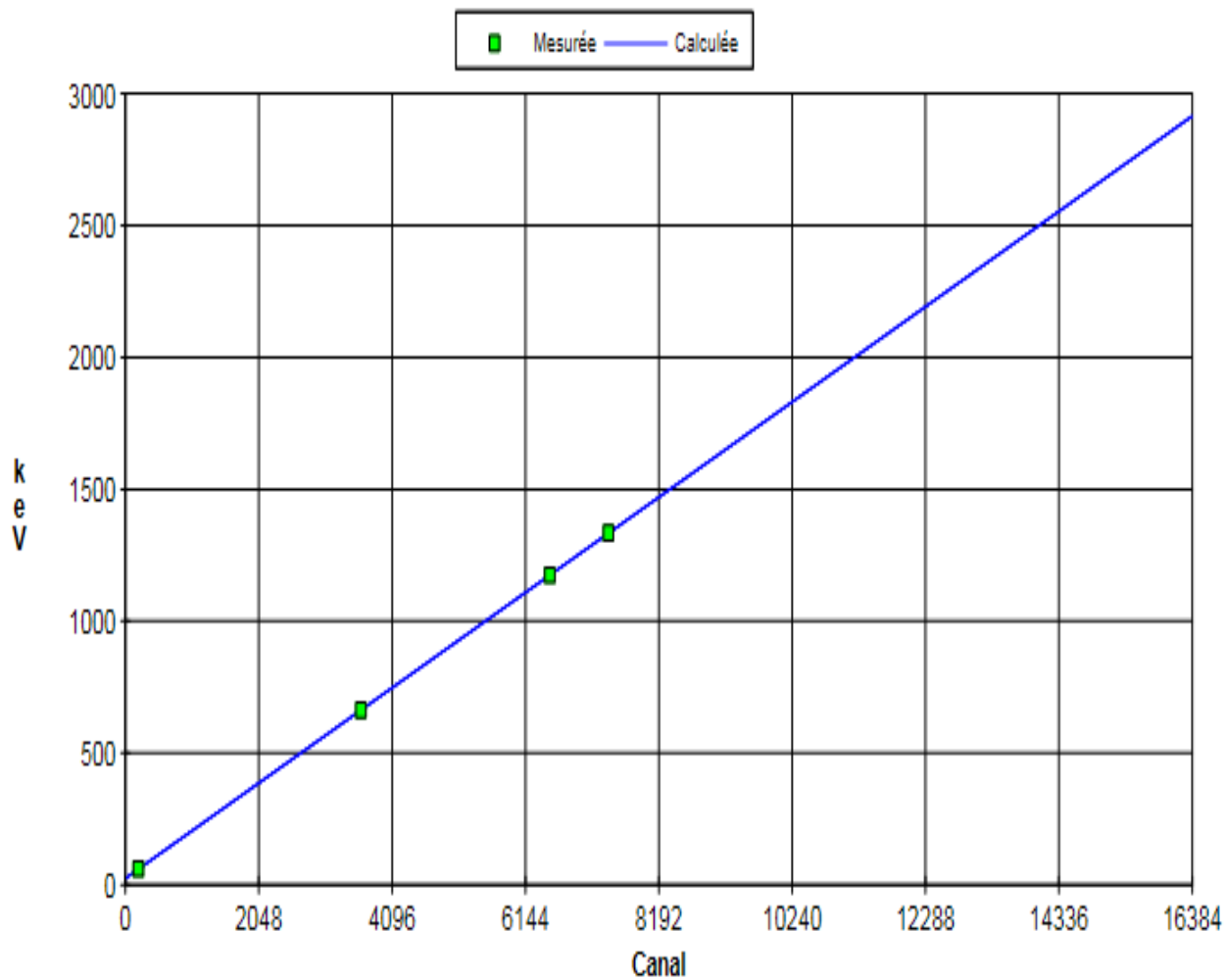


Figure III.4: Energy calibration curve

### 3.3. Efficiency calibration

The calibration of the detector is done using a standard that has the same matrix and same geometry as the measuring samples (figure III.5); it is powder milk contaminated with a radioactive source of Eu-152 with a known activity ( $A_{Eu-152} = 326 \text{ Bq}$  on 06/05/1993) and a half-life of 13.33 years, packaged in a Marinelli beaker of 250 cc size (a container with a curvature in its bottom so the detector would fit inside and have a wider detection range). We place the standard on the detector and we set our chain for a period of 2 h (7200s).



Figure III.5: Standard sample used for calibration in efficiency

After 2h of acquisition, we get the spectrum shown in Figure III.6.

The resulted spectrum shows all the energy peaks that Eu-152 has along with a peak with an energy of 1460.8 keV, the peaks belong to K-40 which is a primordial radio element that appears in every measurement because of its heavily presence in our daily life.

The efficiency of the measuring chain reflects the fact that all gamma photons emitted by the source are not in the direction of the detector, that all gamma photons in the solid angle «Source-Detector»,  $\Omega$ , do not interact within the detector since some can pass through. Moreover, gamma photons that have interacted within the detector have not necessarily yielded all their energy and filled the total absorption peak corresponding to their incident energy. The efficiency of the chain is none other than the detection efficiency; it is the ratio of the number of photons detected to the number of photons emitted by the radioactive source.

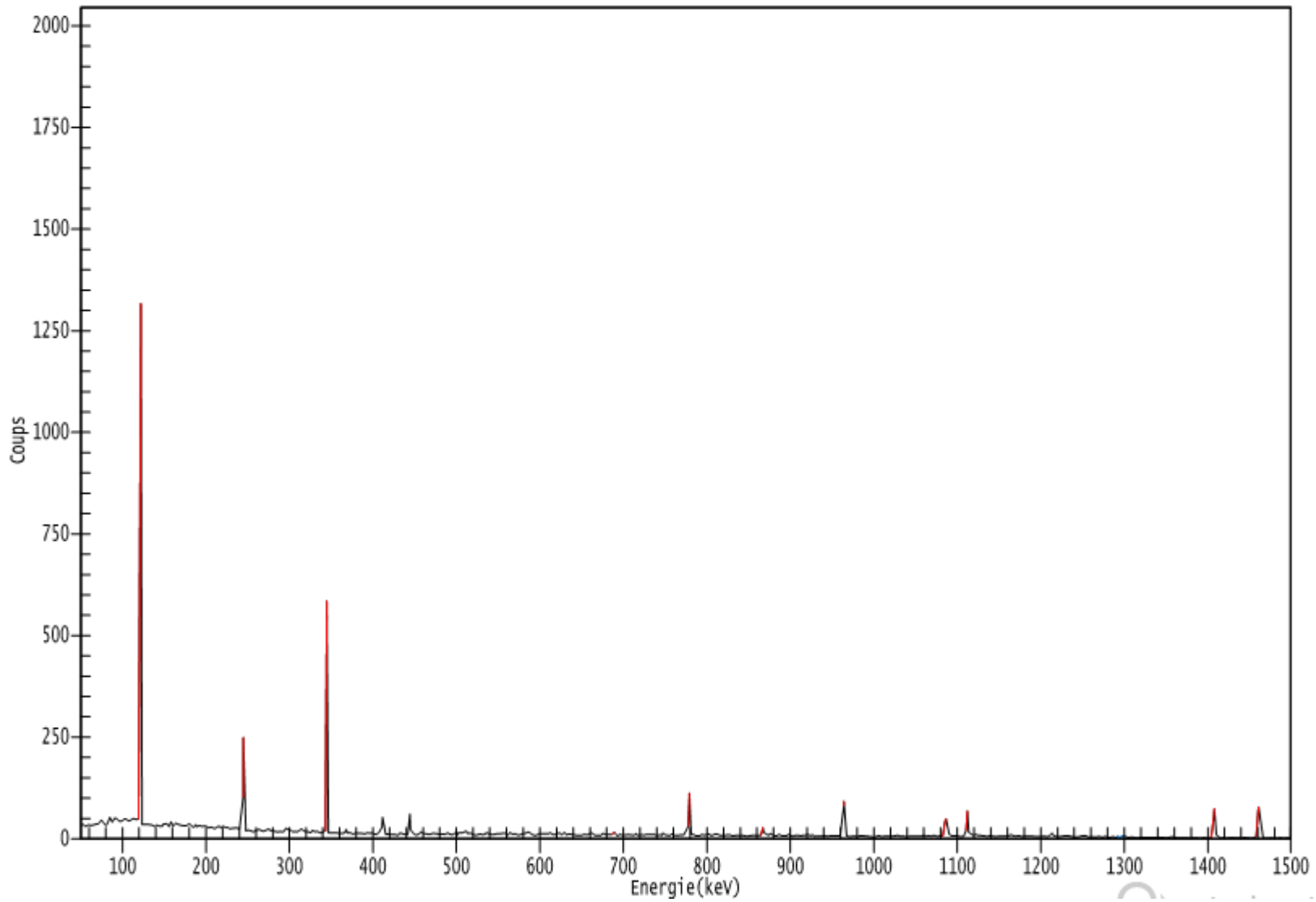


Figure III.6: The standard sample acquisition spectrum



The equation bellow expresses the detection efficiency,

$$\epsilon = \frac{N(E)}{I.A(t).t_c} \dots\dots\dots(III.2)$$

where:

$\epsilon$ : detection efficiency;

N(E): Net area of the characteristic peak (gross area minus background noise), expressed in cps (counts per second);

A(t): Source activity at the time of measurement, in Bq;

I: Probability of gamma-ray emission, in percentages (%);

$t_c$ : Acquisition time, in seconds (s).

To carry this equation out, we must first determine the activity of Eu-152 in the standard the day the acquisition was displayed (20/07/2020). To do so, we use the law of radioactive decay given by the following equation:

$$A(t) = A_0 \cdot e^{-\lambda \cdot t} = A_0 \cdot e^{\frac{-\ln 2 \cdot t}{T_{1/2}}} \dots\dots\dots(III.3)$$

where:

$A_0$ : Activity of the source at the date of manufacture, in Bq ;

t: Time elapsed between the reference date (manufacture) and the measurement date.

All the characteristics of the standard and the calibration results are indicated in the Table III.2.

Source	Energy	N(E) Cps	I (%)	Efficiency
Eu-152	121,8	6759 ±1.29%	28,4	0,042 ±1.14%
	244,7	1330 ±3.28%	7,49	0,031 ±1.81%
	344,3	3969 ±1.66%	26,5	0,026 ±1.29%
	411,1	292	2,56	0,020

		±8.07%		±2.84%
444	332 ±6.98%		3,11	0,019 ±2.64%
778,9	964 ±3.87%		12,74	0,013 ±1.97%
867,4	279 ±7.6%		5,9	0,008 ±2.76
964,1	980 ±3.56%		14,4	0,012 ±1.89
1085,9	556 ±5.52		10	0,010 ±2.35
1112,1	806 ±4.05		13,3	0,011 ±2.01
1408	1045 ±3.25%		20,7	0,009 ±1.80

Table III.2: Efficiency calibration

Figure III.7 shows the efficiency-to-energy variation curve given by the Génie2000 software.

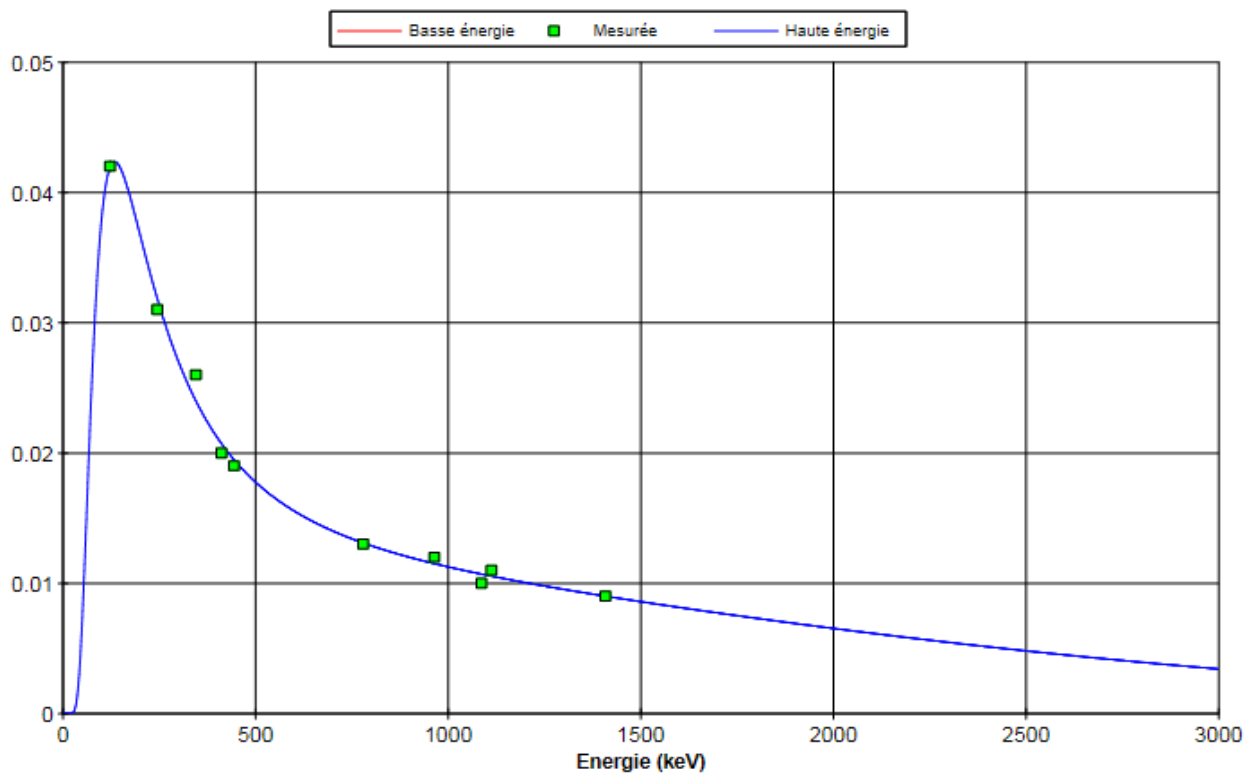


Figure III.7: The efficiency-to-energy variation curve

The equation of the calibration curve given by Génie2000:

$$\ln(Eff) = -1,602 \cdot 10^2 + 1,031 \cdot 10^2 \cdot \ln(E) - 2,497 \cdot 10^1 \cdot \ln(E)^2 + 2,651 \cdot 10^0 \cdot \ln(E)^3 - 1,048 \cdot 10^{-1} \cdot \ln(E)^4 \dots\dots\dots(III.4)$$

This equation allows us to quantify the activity of any radio element that makes an appearance in our measurement spectrum by its detection efficiency.

#### 4. Detector characterization

##### 4.1. Energy resolution

Energy resolution measures the ability of a detector to separate two adjacent energy lines. The Full Width at Half Maximum (FWHM) resolution, expressed in keV, is the full width of the photoelectric peak measured at mid-height. Indeed, a precise measurement of two consecutive lines becomes possible when the energy difference E is less than the FWHM.

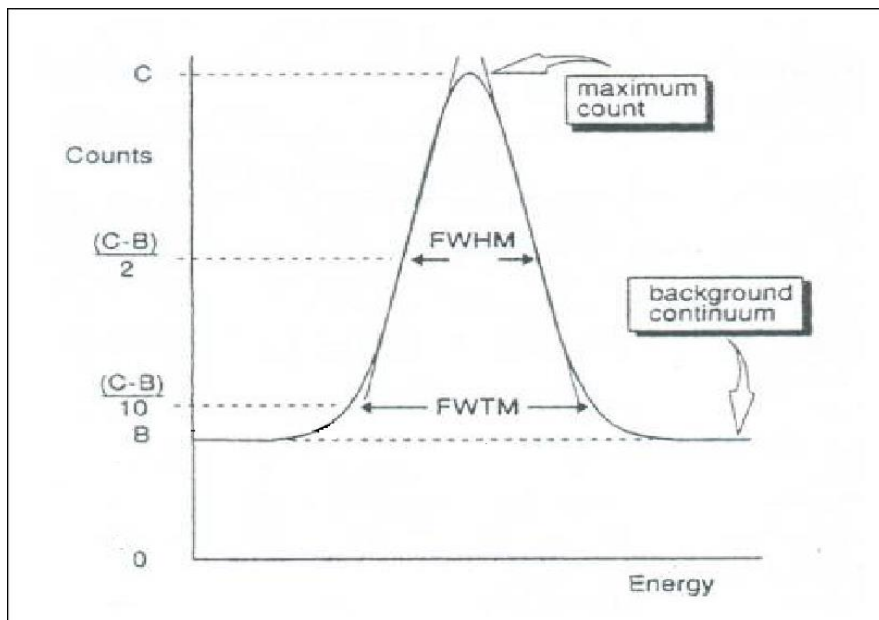


Figure III.8: Resolution definition of a perfectly Gaussian peak

Experimentally, the R resolution (expressed in %) of the Ge(HP) detector being the ratio of the FWHM resolution to the energy of the second peak of Co-60 (1332.5 keV):

$$R(E) = \frac{FWHM}{E} \times 100 \dots\dots\dots(III.5)$$

The FWHM value is given by the Génie2000; in our case it is 2.344 keV

$$R(E) = \frac{2.344}{1332.5} \times 100 = 0.17\% \dots\dots\dots(III.6)$$

**4.2. Detection limit and minimum detectable activity**

The detection limit ( $L_d$ ) sets the threshold below which the measuring equipment is no longer sensitive enough for the sample in question. This is the lowest value of a quantity that can be measured, so it expresses the sensitivity of the measuring technique. For each energy peak found, a detection limit is calculated. To do so, you must first pass through the detection limit ( $L_c$ )(Génie2000 n.d.). That is defined as:

$$L_d = k^2 + 2L_c \dots\dots\dots(III.7)$$

with:

$$L_c = k \cdot \sqrt{F + \sigma_F^2} \dots\dots\dots(III.8)$$

where:

$L_d$ : Threshold of detection;

$L_c$ : Detection limit;

k: Confidence factor (coefficient of error), is equal to 5%; k= 1,645 corresponding to a 95% confidence level, for the calculation of AMD ;

$\sigma_F^2$ : Error on continuous background (background noise);

F: Background noise of the area of interest (total area).

The value of F is taken from the background noise spectrum after the marking of the region of interest of each radionuclide searched.

where:

$$\sigma_F = \sqrt{F} \quad \longrightarrow \quad \sigma_F^2 = F$$

This makes it possible to write the following:

$$L_c = k \cdot \sqrt{2F} \dots\dots\dots(III.9)$$

If we replace the equation (III.9) in equation (III.7), we can write:

$$L_d = k^2 + 2k \cdot \sqrt{2F} \dots\dots\dots(III.10)$$

We also mentioned that k=1.645 which gives us:

$$L_d = 2.71 + 4.65 \cdot \sqrt{F} \dots\dots\dots(III.11)$$

The Minimum Detectable Activity (MDA) is ultimately the conversion of the detection limit ( $L_d$ ) to activity (in Bq). It is calculated by the following relation:

$$MDA = \frac{L_d}{\epsilon \cdot I \cdot t} \dots\dots\dots (III.12)$$

Since the only radio element that made an appearance in our spectrum along with Eu-152 is K-40, we should count its detection limit and minimum detectable activity, where:

$$F = 90 \text{ cps} \quad \text{and} \quad k = 1.645$$

Thus, we get:

$$L_d = 46.82 \text{ cps} \quad \text{and} \quad MDA = 5.7 \text{ Bq}$$

### 5. Measurement of radioactivity in milk powder samples

Samples to be analyzed are a milk powder from 13 different origins: Canada, Poland, United States of America (USA), Uruguay, New Zealand, Ukraine, Spain, France, Holland, Ireland, United Kingdom (UK), Argentina and Lithuania. The samples are put in Marinelli beakers of 250 cc size.

#### 5.1. Background acquisition

For better measurement accuracy, we launched background acquisition for 2h (7200 s) with an empty Marinelli beaker of 250 cc size, so we can latter subtract it from the area of the interest. The resulted spectrum is shown in Figure III.9.

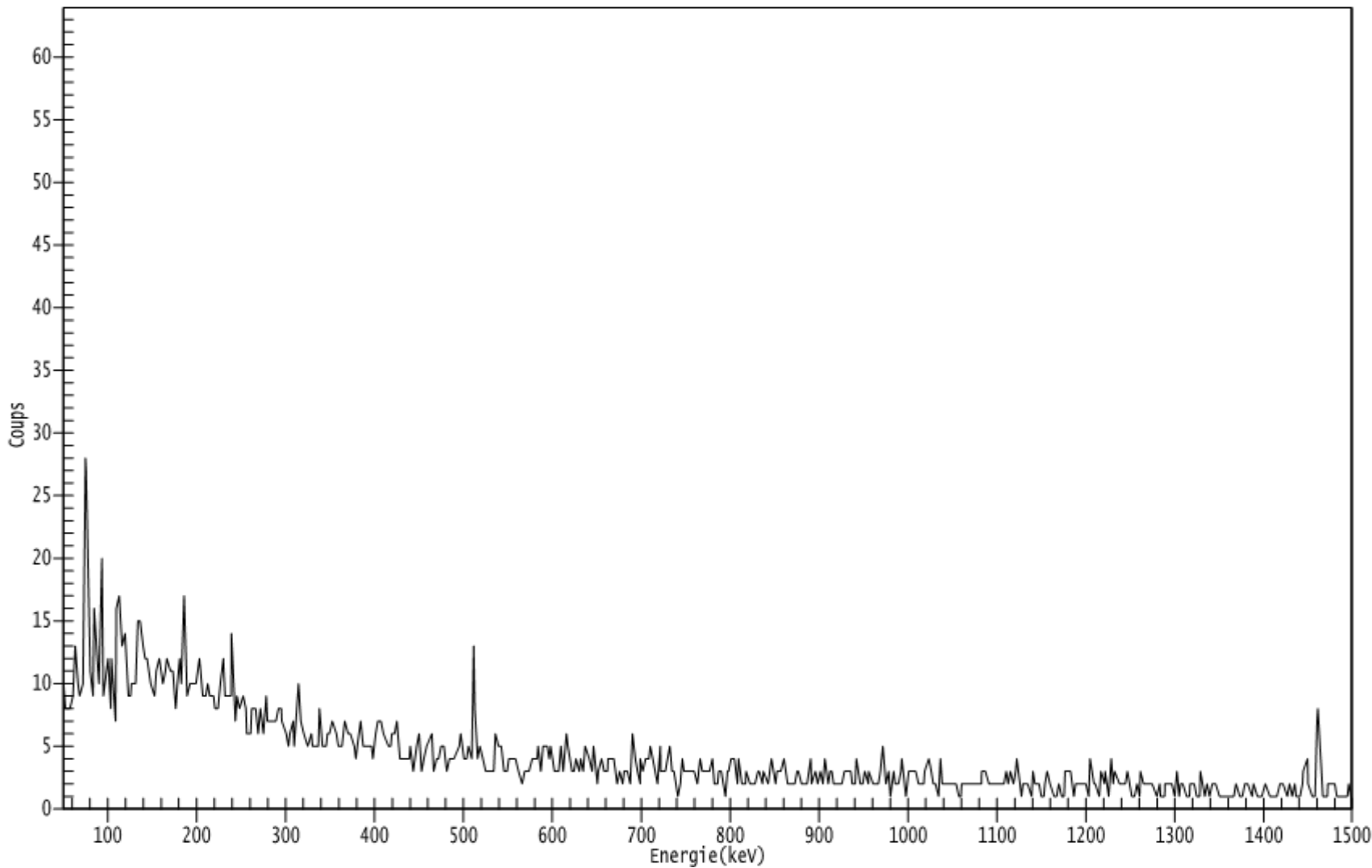


Figure III.9: Background spectrum

### 5.2. Qualitative analysis

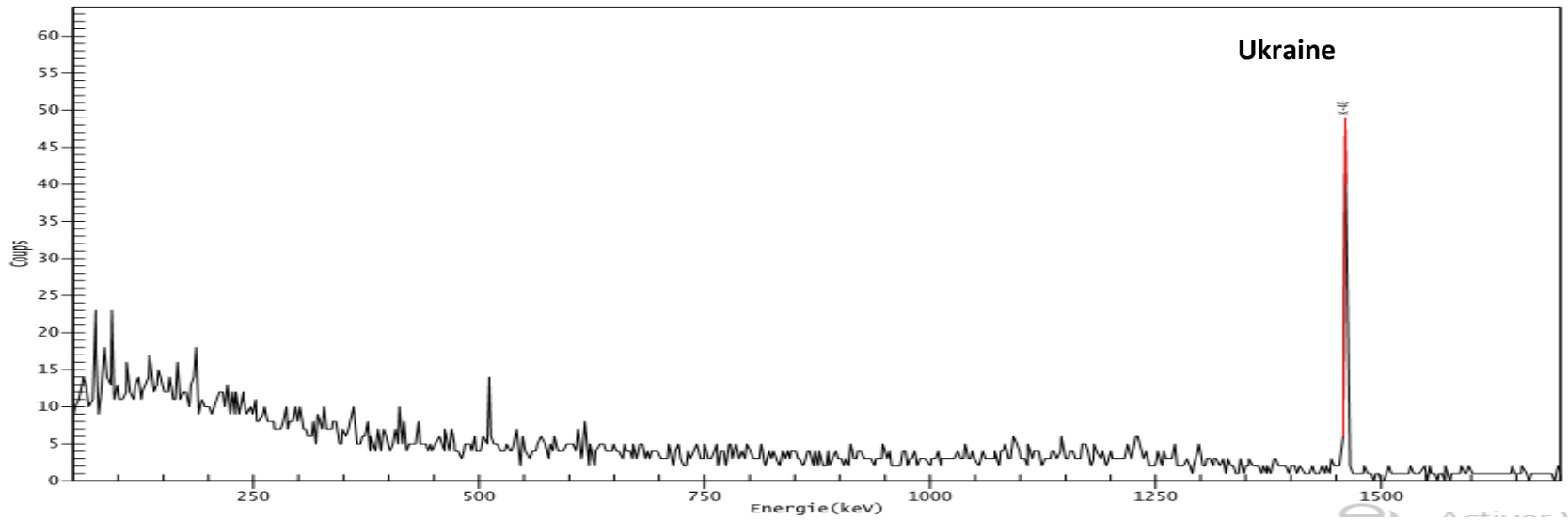
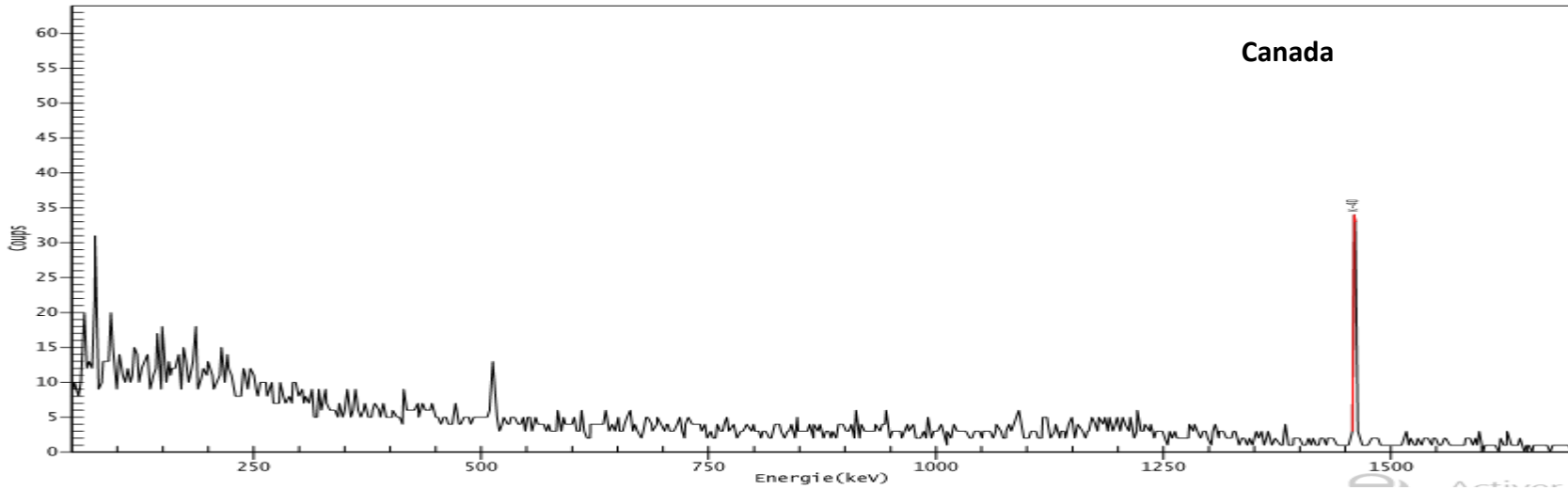
This analysis consists in defining the zones corresponding to peaks of total absorption whose surface will allow matching energies. These areas are referred to as “Regions of Interest: ROI”. Thanks to a library of radioisotopes supplied with the acquisition software, it is possible to identify the radionuclides present in the sample just by knowing the energy of the total absorption peaks.

After preparing the samples to be analyzed, conditioning it in geometry identical to that of the calibrated standard and placing them in the detector one by one, we launch the acquisition. For each sample, the scan time is set to 7200s. Each spectrum is recorded and saved by Génie2000.

### 6. Acquisitions results

Figure III.10 illustrates the spectrums obtained from the measurements of the samples from Canada, Ukraine and France respectively.

The remaining spectrums are illustrated in figures A.1, A.2, A.3, A.4, A.5, A.6, A.7, A.8 and A.9) (View annex).





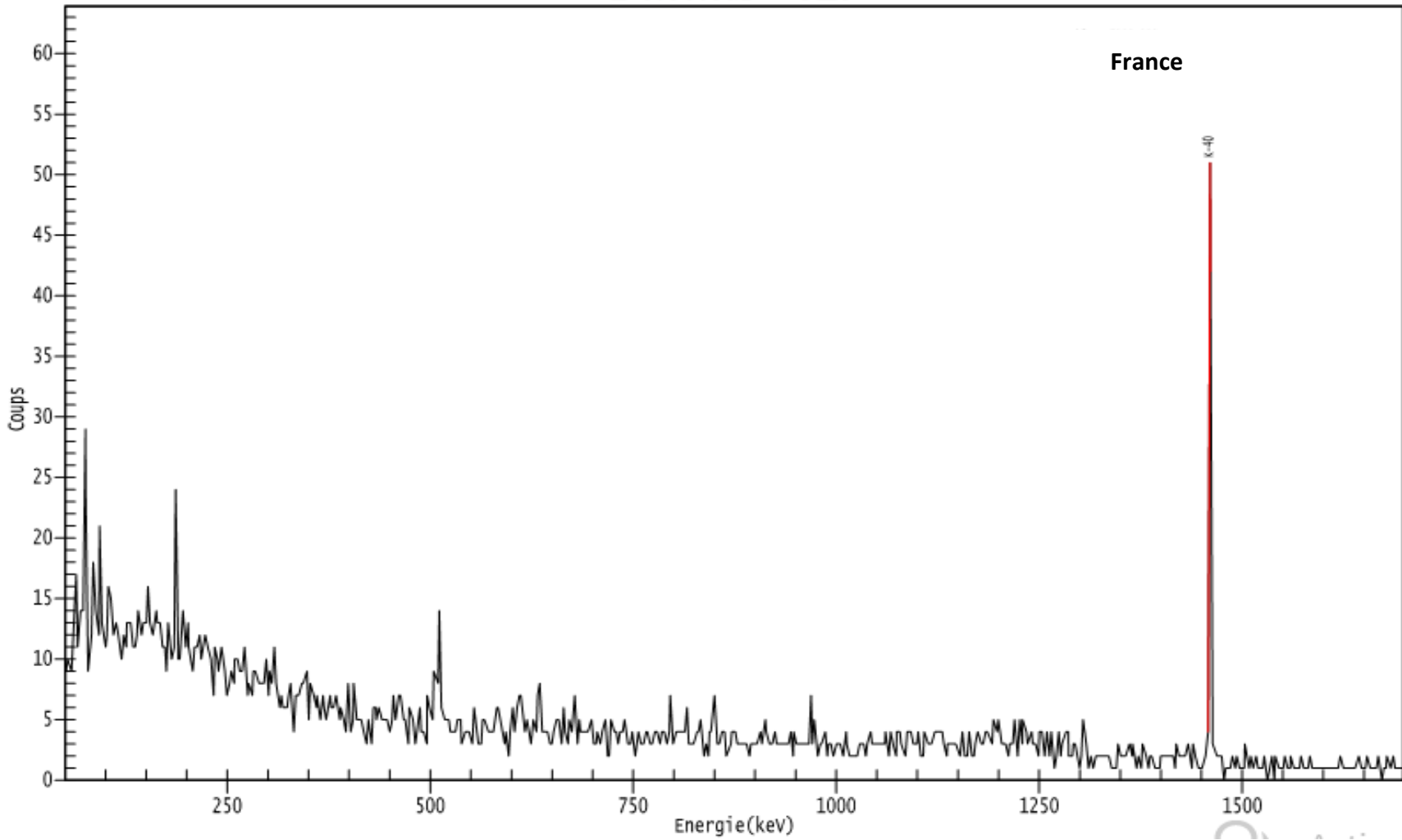


Figure III.10: Spectrums obtained from the measurement of power milk samples from Canada, Ukraine and France respectively

After the qualitative analysis we observed the presence of K-40 in all milk powder samples, with an energy peak of 1460.8keV and an intensity of 10.67%.

**Chapter IV : RESULTS AND DISCUSSION**

Once we are done with the samples analysis and got the spectrums that are illustrated in chapter four, we get next to interpret and discuss them.

The previous chapter indicates that the only radionuclide present in all of the 13 samples is K-40, which is a natural radionuclide that doesn't present any contamination risks.

For the sake of this work we will quantify the concentration of K-40 in the samples eventhough it does not make a threat.

**1. Activity quantification**

The K-40 activity in the samples was calculated using the following formula:

$$A(t) = \frac{N(N_{gross}-N_b)}{\epsilon.I.t} \dots\dots\dots(IV.1)$$

*N<sub>gross</sub>*: Gross area of the interest peak from the sample spectrum.

*N<sub>b</sub>*: Area of the interest peak from the background noise spectrum.

The associated uncertainties are given by the equation:

$$\frac{\Delta A}{A} = \sqrt{\left(\frac{\Delta N}{N}\right)^2 + \left(\frac{\Delta \epsilon}{\epsilon}\right)^2 + \left(\frac{\Delta t}{t}\right)^2 + \left(\frac{\Delta I}{I}\right)^2 + \left(\frac{\Delta m}{m}\right)^2} \dots\dots\dots(IV.2)$$

where:

m: the sample weight, in (g) ;

$\Delta m = 0.01 \text{ g}$  ;

$\Delta t = \Delta I \cong 0$  ;

The maximum value of uncertainty on efficiency is set at 3%;  $\Delta \epsilon = 3\%$  where the actual value of efficiency was calculated in chapter IV (  $\epsilon = 0.0107$ ).

By having the activity of K-40, we can also define the specific activity *A<sub>s</sub>* (Bq/kg).

$$A_s = \frac{A(t)}{m} \dots\dots\dots(IV.3)$$

Samples number	Origin	Gross area (cps)	$A_{K-40}$ (Bq)	$As_{K-40}$ (Bq/kg)
1	Canada	724 ±3,81	79,20 ±4.85	316,78
2	Poland	643 ±4,06	69,34 ±5.05	277,37
3	USA	662 ±3,97	71,65 ±4.98	286,61
4	Uruguay	404 ±5,09	40,27 ±5.91	161,07
5	New Zealand	419 ±4,99	42,09 ±5.82	168,37
6	Ukraine	614 ±4,04	65,81 ±5.03	263,25
7	Spain	573 ±4,24	60,83 ±5.19	243,30
8	France	679 ±3,89	73,72 ±4.91	294,88
9	Holland	653 ±4,02	70,56 ±5.02	282,23
10	Ireland	446 ±4,93	45,38 ±5.77	181,50
11	UK	583 ±4,21	62,04 ±5.17	248,17
12	Argentina	443 ±4,95	45,01 ±5.79	180,04
13	Lithuania	520 ±4,46	54,38 ±5.38	217,51

Table IV.1: Calculation of activity and specific activity of K-40 in samples

## 2. Results interpretation

Based on the qualitative analysis of all the spectrums obtained for the 13 milk powder samples from different origins, we can note the following:

- The absence of artificial radioactivity in all analyzed samples, which means the non-radio contamination of the samples, which approves their quality and their safety for human consumption.
- The radioactivity present in the thirteen analyzed samples represents the natural radioactivity and is due essentially to radionuclide potassium-40 (K-40).
- We can observe that the K-40 activity differs from a sample to another; the obtained values slightly vary between 79.20 Bq and 40.27 Bq. We can also see that powder milk originating from Canada holds the record for the highest K-40 activity concentration

followed by France and USA, while Uruguay holds the lowest K-40 activity concentration.

It is important to mention that powder milk is initially made from liquid milk, which comes out of cows. Those last feed on grass and water. Having a higher activity in Canada, France and USA, which are considered producer leaders, raises questions about their raw resources conditions.

### 3. Recommendations

To finalize this work, we would like to make some recommendations to the managers in order to improve the quality of service provision:

- The received samples are brought and delivered by importers. This clears paths for frauds to cheat their ways to the non-contamination certificate by changing the samples with others on their way to the service. The samples should be delivered by the veterinary service that is in charge of sampling imported products.
- The sampling in the ship should follow a specific and normalized process. This is done by people from other institution (veterinary service). We have no warranty about the respect of the process. It would be better if people from the service realize the task.
- The number of demands is increasing daily thus the number of samples. Meanwhile, the service team is composed of only three members, therefore it is necessary to empower the team by adding more people.
- Some of the gamma spectrometry chains are old and need to be replaced.

## Conclusion

The present work focused on the qualitative and quantitative analyses of the levels of radioactivity present in imported milk powder, in order to ensure that they are not radioactive and are safe for human consumption.

We have chosen to analyze samples of milk powder imported from different producer countries. For this, we settled down with the gamma spectrometry technique using a Hyper Pure Germanium detector Ge(HP). This non-destructive measurement technique is considered to be a very powerful and responsive technique with a high resolution. Thus, it makes it easy to identify and determine the activity concentration of any natural or artificial radionuclide present in samples. The technique also supports the diversity in samples matrixes and geometry which makes it a better fit and suitable for foodstuff analysis.

Before carrying out the qualitative and quantitative analysis of the different milk powder samples, we carried out first a calibration in terms of energy and another in the efficiency of the measuring chain. This calibration is needed to assume the required precision and

Based on the qualitative analysis of thirteen milk powder samples, we noted the absence of any artificial radioactivity in all analyzed samples.

However, we also noted the presence in all samples of K-40 as a natural radionuclide. Regarding quantitative analysis, we noted that the activity of K-40 is relatively higher in samples from certain countries that are considered leaders in milk powder production (Canada, France and USA), than in countries that are less productive (Uruguay). This is probably due to the raw resources conditions.

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ANNEX

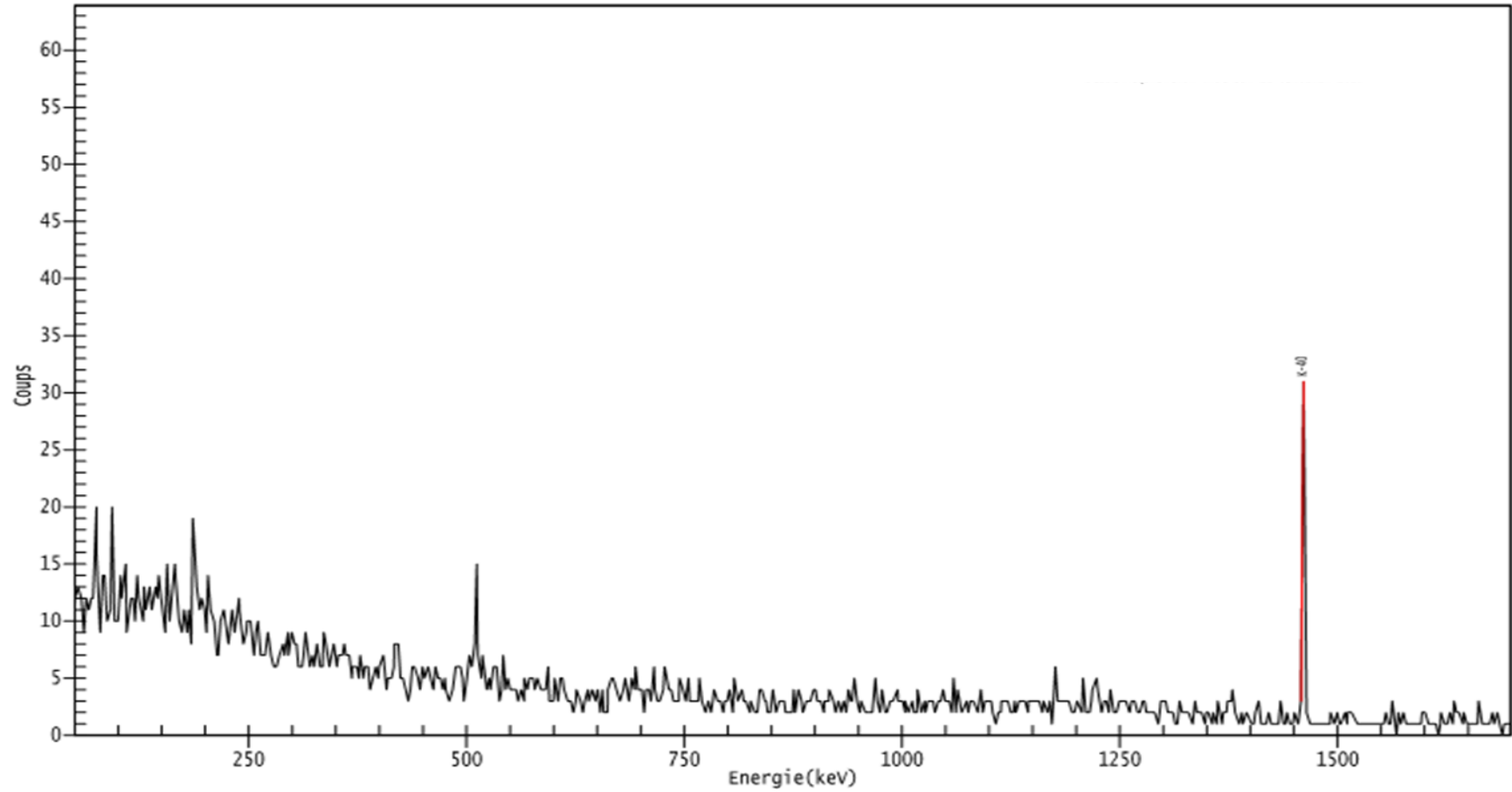


Figure A.1: Spectrums obtained from the measurement of power milk sample from Argentina



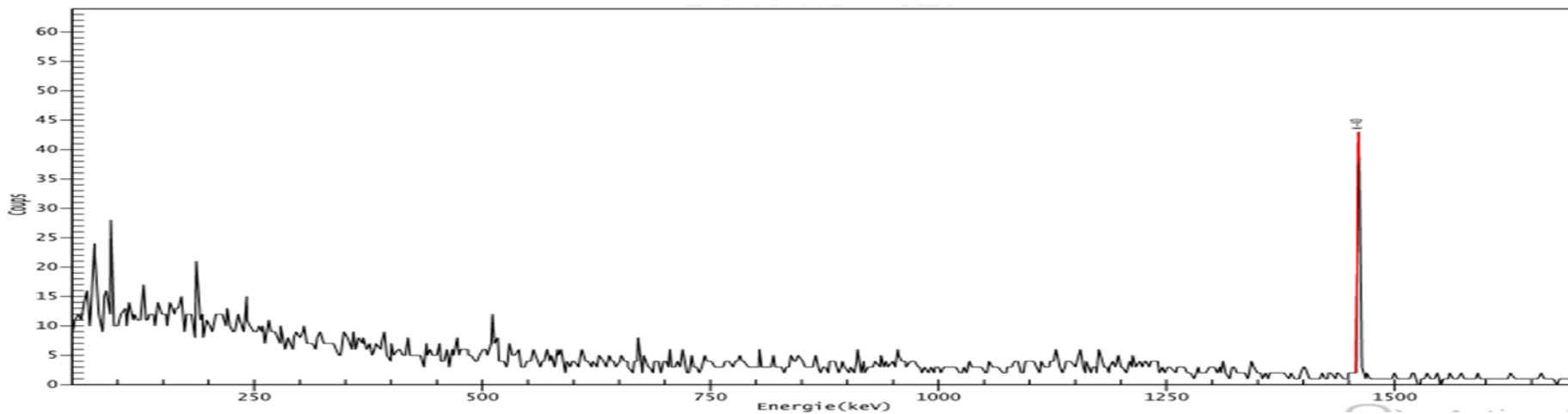


Figure A.2: Spectrums obtained from the measurement of power milk sample from Holland

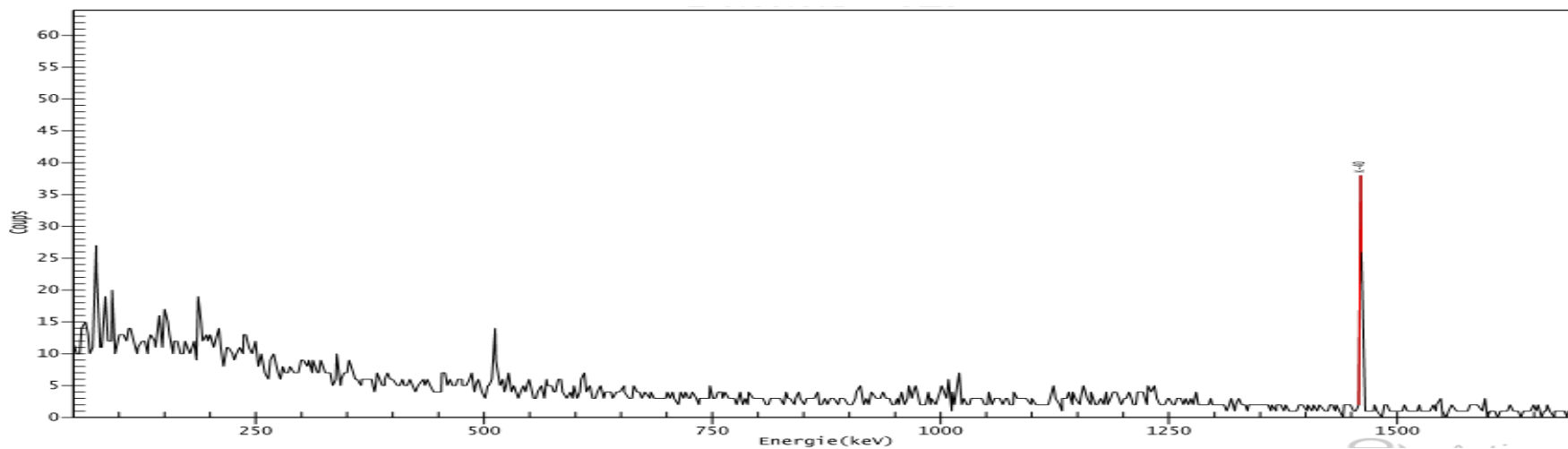


Figure A.3: Spectrums obtained from the measurement of power milk sample from Ireland

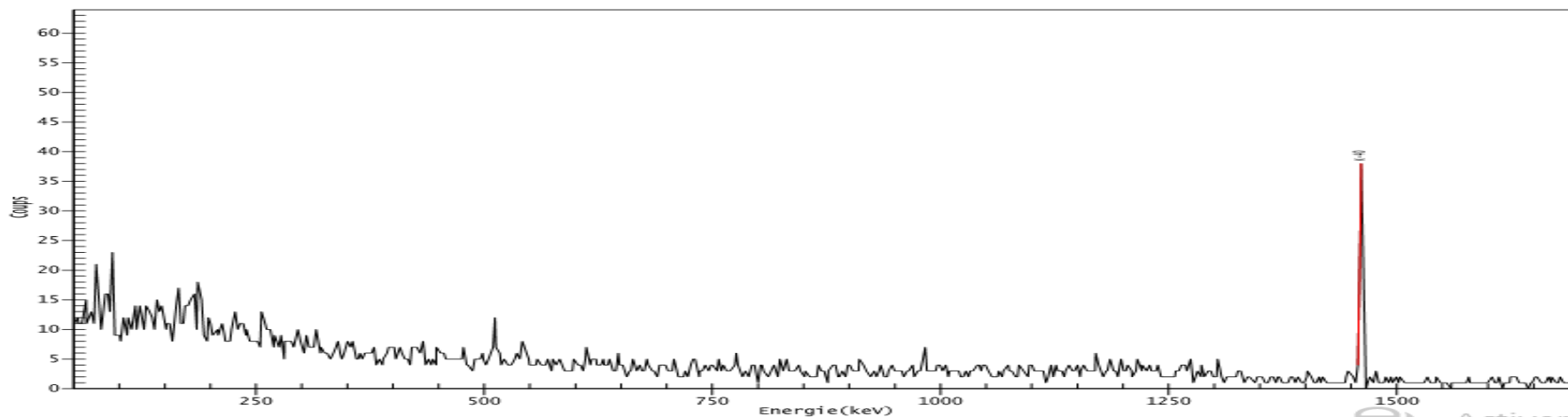


Figure A.4: Spectrums obtained from the measurement of power milk sample from Lithuania

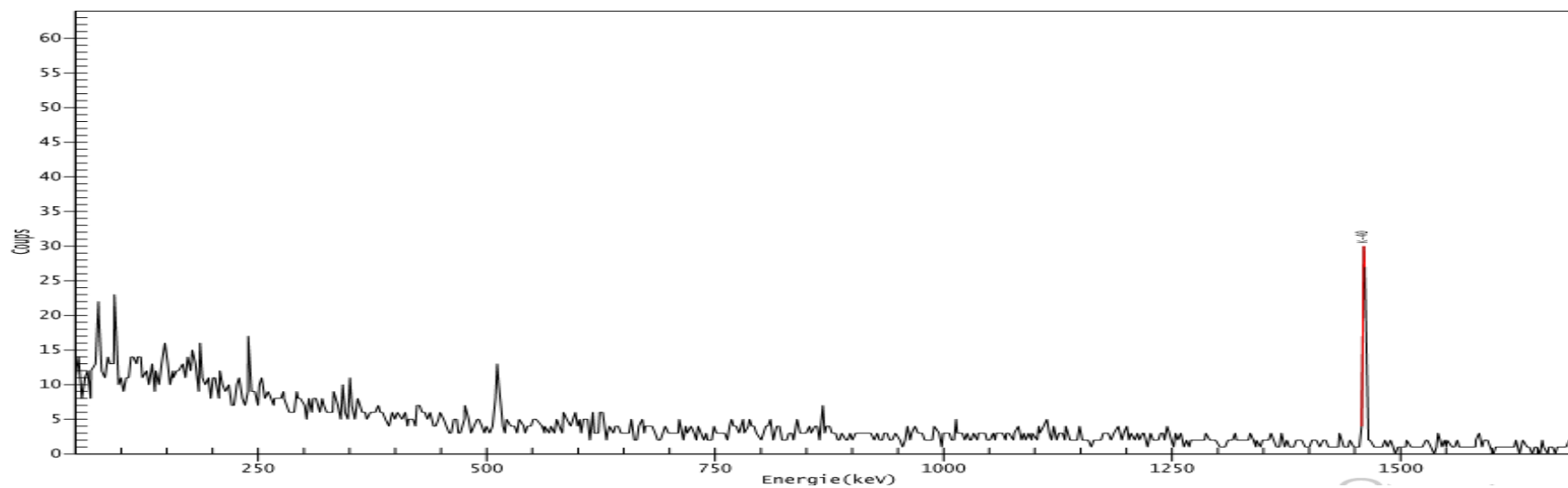


Figure A.5: Spectrums obtained from the measurement of power milk sample from New Zealand

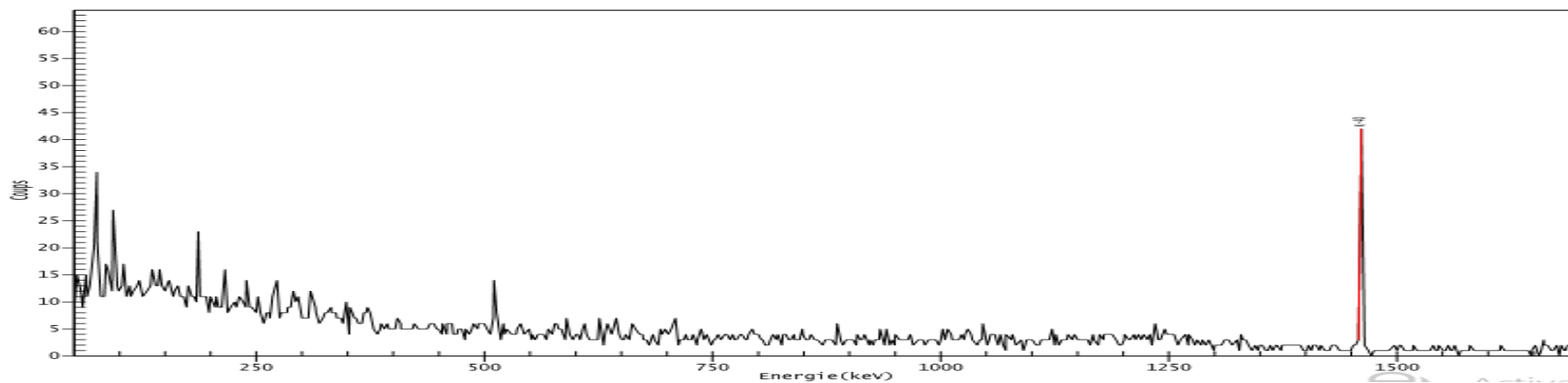


Figure A.6: Spectrums obtained from the measurement of power milk sample from Poland

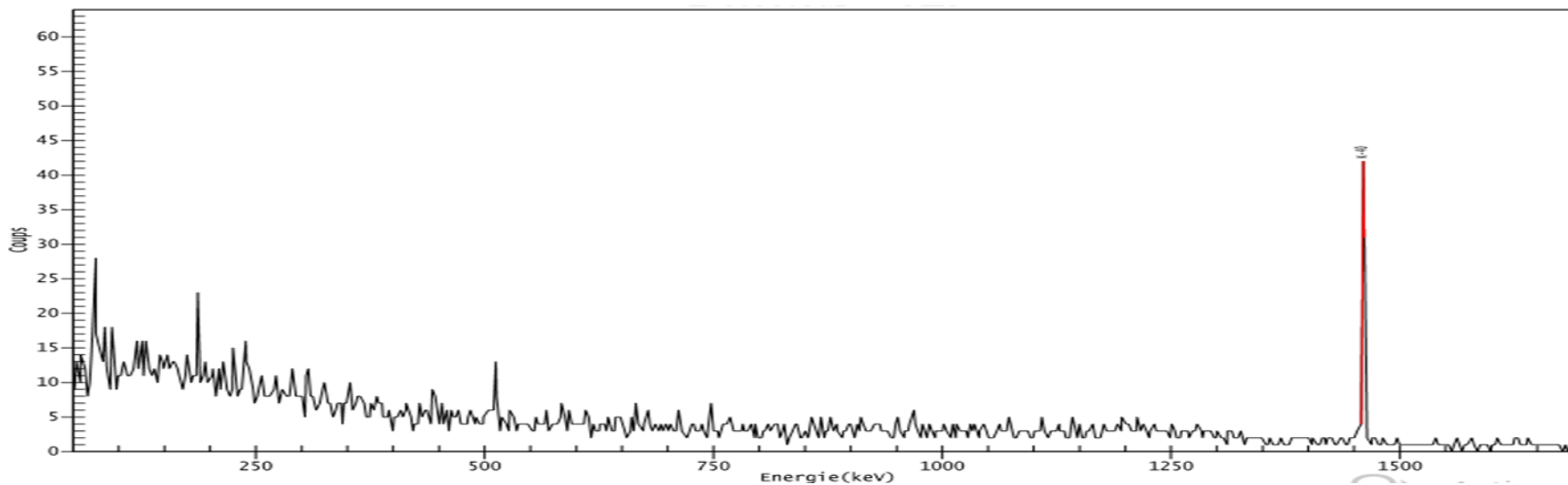


Figure A.7: Spectrums obtained from the measurement of power milk sample from Spain

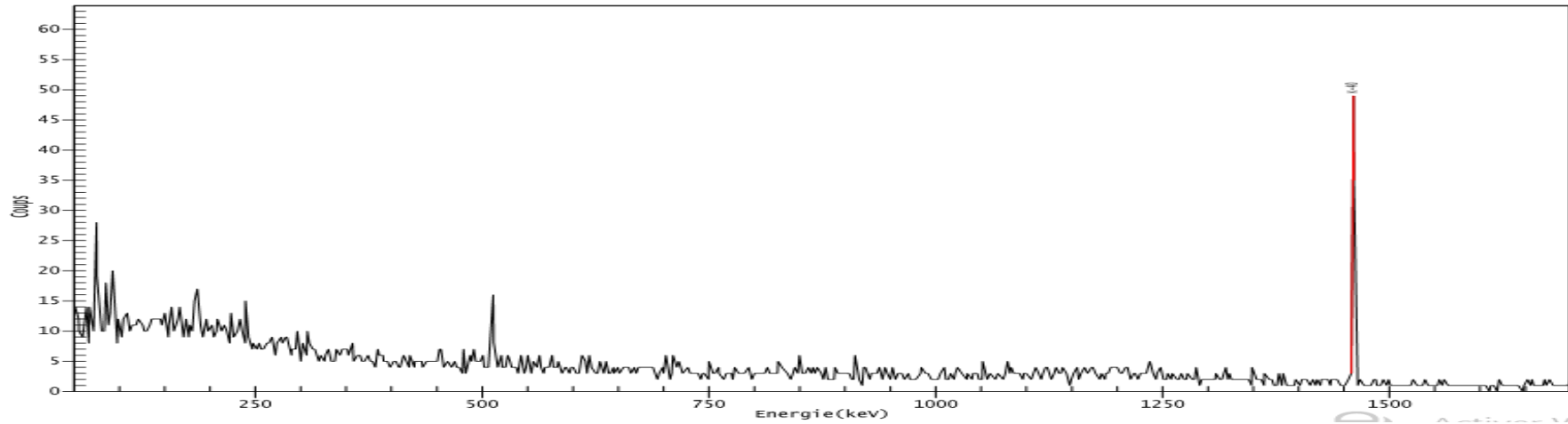


Figure A.8: Spectrums obtained from the measurement of power milk sample from U.K

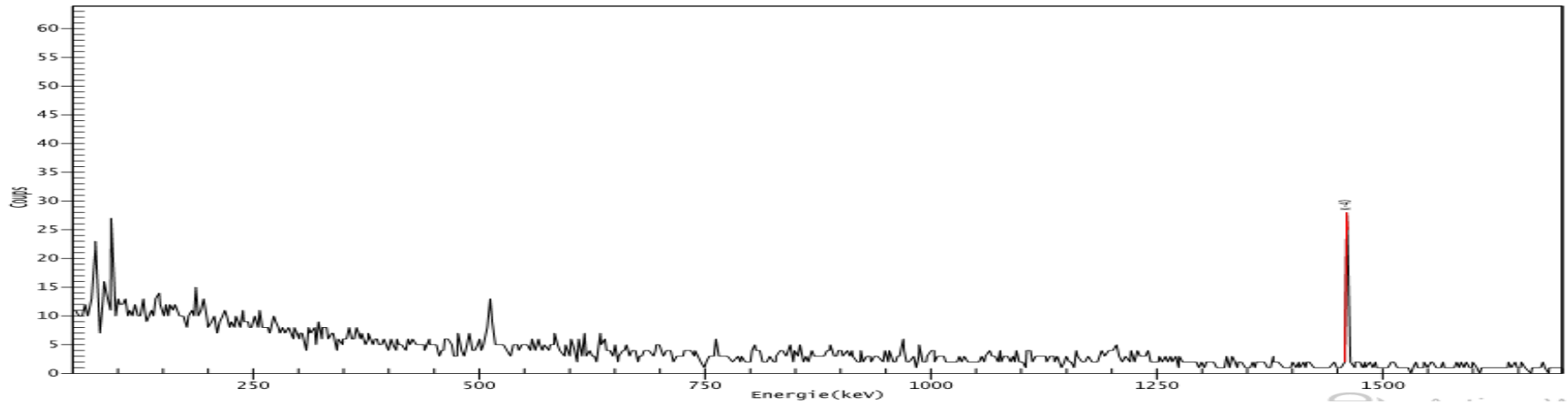


Figure A.9: Spectrums obtained from the measurement of power milk sample from Uruguay

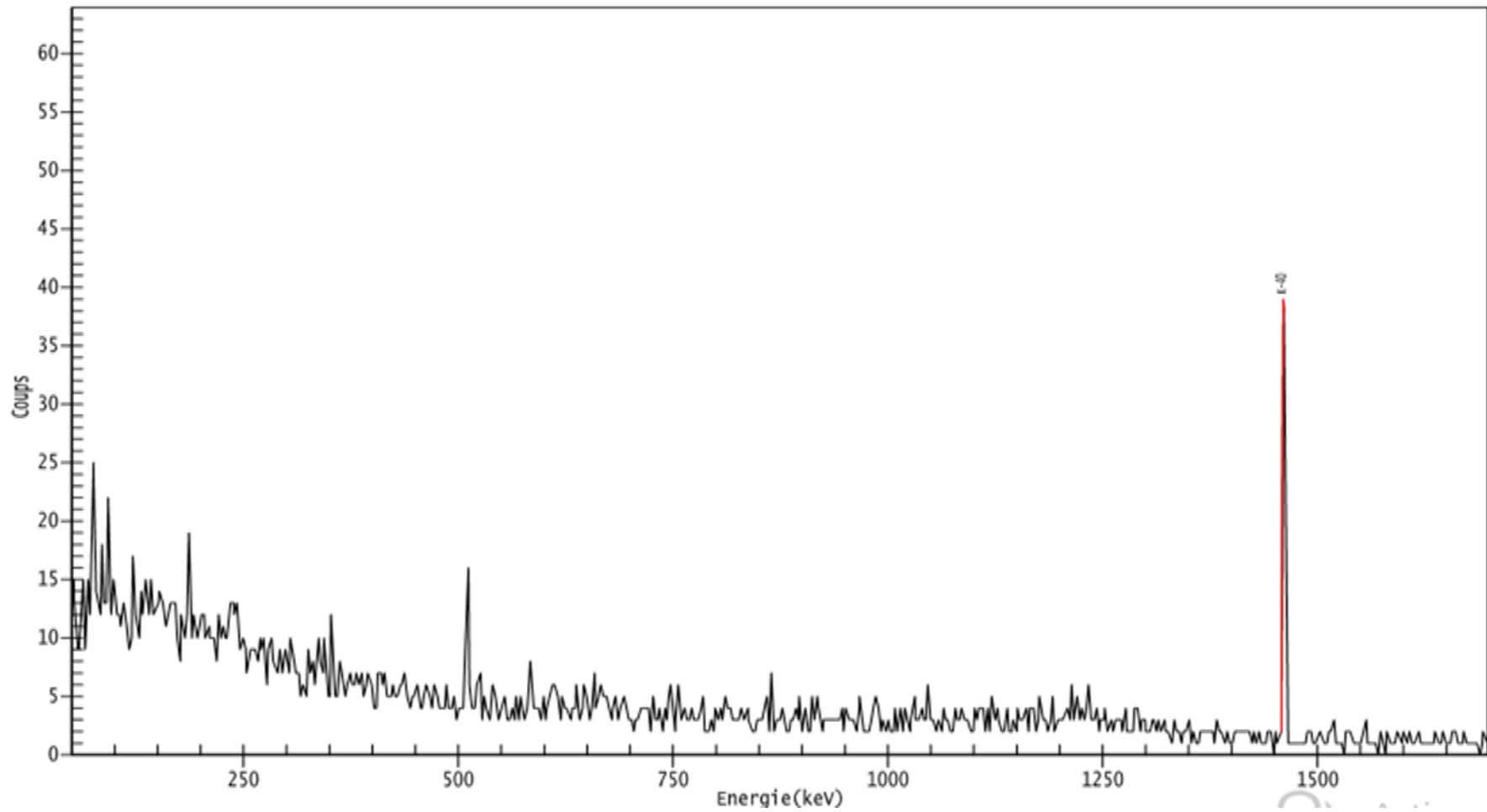


Figure A.10: Spectrums obtained from the measurement of power milk sample from USA