Method for the identification of ⁶⁰Co in an industrial gamma irradiator

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This study presents a method for the identification of a set of radioactive sources of 60 Co, in an industrial irradiator. This identification was requested by a Mexican regulatory authority. To fulfill this request, a method was developed to make the identification by the gamma spectrometry technique. The problem for the identification of Co-60 lies in the fact that the source (set of Co-60 rods) has a total radioactive activity of two million Ci, so the gamma fluence rate is very high and saturates the gamma system, preventing the corresponding photopeaks in the gamma spectrum to be appreciated. A strategy was applied to reduce saturation, pile up, and dead time to ensure that the photopeaks generated in the 60 Co gamma energy spectrum were clearly visible. A low efficiency and relatively high resolution CdZnTe detector, was selected, which was placed in an acrylic box with a collimated lead shield to carry out the measurement inside the irradiator pool to reduce scattered radiation and to be able to clearly appreciate the spectrum gamma of 60 Co.

Keywords: Gamma spectrum; pileup; industrial irradiator; CdZnTe.

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1. Introduction

Industrial Gamma Irradiators are widely used in the world, they are used to sterilisation, innocuity and preservation of food products among other applications. Generally, this type of irradiators are composed of radioactive sources of ⁶⁰Co. To achieve the absorbed dose necessary for the irradiation of products, there are industrial gamma irradiators with typical activities of hundreds of thousands Curies (Ci) of radioactivity.

In particular, the gamma irradiator to which it was requested to identify ⁶⁰Co has an activity of two million Ci. Because ⁶⁰Co is a beta-gamma emitter and it emits photons with 1.17 and 1.33 MeV of energy with yields of practically 100%; the gamma spectroscopy method (GS) was used to make the measurement, which is based on the interaction of gamma radiation with matter by photoelectric effect. Thus, in an instrumented GS system, the electrical pulse is of the same amplitude when the interaction of gamma photon with the detector is by this effect and this fact allows to identify gamma emitter radionuclides by its energy.

The result of a counting by gamma spectrometer is a gamma spectrum which is a histogram composed of frequency of events (photoelectric, Compton scattering and pair production) where the photopeaks of certain energies stand out and that allow to identify gamma emitting radionuclides. Various types of detectors are used in GS systems, the simplest being scintillators and the most modern being semiconductor detectors.

Basically, the identification of radionuclides by GS is a simple process without any complications, but in this case the high intensity of the source constitutes a problem to carry out the identification. Since the main problem to perform the measurement was the high intensity of the source, it was decided to use a spectrometry detection system that met three conditions: a) Low efficiency b) Good resolution c) Portability. For this reason, it was decided to use a system with a modern CdZnTe semiconductor detector, which met the above conditions and with which the identification of the source was made.

2. General instrumentation

A gamma spectrometry system basically consists of: a) Gamma Radiation Detector b) Power Supply c) Preamplifier d) Amplifier e) Multichannel Analyzer (MCA) f) Shielding. The detector is the main part of the system and it is generally a scintillation or semiconductor detector that works under the following mechanisms. When gamma radiation interacts with the detector, positive and negative ions in the form of ejected electrons and ionized atoms respectively are generated by photoelectric, Compton and pair production effects, these ions are collected and processed in the following stages.

The power supply provides the potential difference (within the sensitive volume of the detector) necessary to produce the electric field that directs the ions to the electrodes (or dynodes in a scintillation detector) and be processed by the preamplifier to direct them to the amplifier.

The preamplifiers' main purpose is to provide of impedance coupling and to transform the electric charges into a voltage pulse which in turn is directed to the amplifier input. The amplifier shapes the electronic pulse from the preamplifier and, depending on the type of output selected, produces an electronic signal of Gaussian or triangular shape to give the electronic pulses amplitude, according to the range of energy that it is useful to handle.

The multichannel analyser (MCA) is an electronic device that consists of an input window (height of voltage pulses window), an analogue-digital converter (ADC), which "mea-



FIGURE 1. Gamma spectrum containing photopeaks with energies from 59.5 keV to 1836.0 keV for energy calibration.

sures" the pulse amplitude and sends it to a memory location called channel, (typically 2k to 8K channels) in which the number is directly proportional to the output electronic pulse amplitude. The MCA contains a screen (often the monitor of a P.C.) or its own screen, where is possible to see a chart of the distribution of events of different energies recorded (height of voltage pulse), called gamma spectrum, (Fig. 1).

When a gamma photon interacts with the detector by photoelectric effect, the amplitude of the pulse is directly proportional to the energy of the gamma photon and therefore when this type of interactions are repeated for each energy they give rise to "Gaussian-like" distributions called photopeaks. The location of these photopeaks allows the energy to be related to the channel number of the centre of the mentioned distribution, giving rise to the energy calibration, which allows knowing the energy of unknown photopeaks due to their location in the spectrum and therefore the radionuclide emitting that gamma photon.

The shield containing the detector is usually made of lead with different wall thickness in function of the application (for instead, for environmental analysis a thickness of 10 cm is typical) and its function is to prevent background gamma radiation from walls, ceiling floor, and surroundings reaching the detector. Figure 2 shows the basic components of a traditional gamma spectrometer system.







FIGURE 3. Electronic pulses 1, 2 and 3 are the product of three single interactions of photons in the detector, the sum pulse is product of two single interactions 4 and 5, sum peak is product of pileup.

3. Electronic pulse formation

In semiconductor detectors, the holes (positive ions) and electrons produced by gamma radiation contribute to the pulse formation. The mobility of the electrons and holes depends on material characteristics, strength of the electric field applied, and operating conditions. Typically, the charge carrier mobility is on the order of $10^3 - 10^4$ cm²/(V.s). Therefore, for a typical semiconductor detector the charge collection time is about a fraction of a microsecond. If a second event takes place before all the charge from the first event is collected, the charge carriers produced by the second event will be added to the pulse produced by the initial event, hence leading to the phenomenon of pileup, Fig. 3. The time that must elapse between two pulses produced so that no pileup occurs is called the resolution time.

The pileup phenomenon can occur when two gamma photons of different energies come from two different sublevels from the same original energy level in the same nuclear decay process and interact with the detector almost at the same time. Another case is when two photons from different nuclei interact with the detector practically at the same time.

4. Used equipment

The gamma spectrometry system that was used to perform the measurement is a Kromek brand integrated system with the following characteristics and specifications: Model: GR05+; Serial number: 150005; Firmware version: 1.0; Detector size: $10 \times 10 \times 10$ mm; CZT (CdZnTe) detector Energy range: 25 keV to 3.0 MeV; Energy resolution: < 2.5% (661.66 keV, 137 Cs); Maximum throughput: 30,000 counts/s; Number of channels: 4096; Dimensions: $25 \times 25 \times 62$ mm (Fig. 4); Weight: 60 grams. Energy output: Rise time: 3 microseconds; Decay time: 10 microseconds. Timing output: Shape: TTL compatible rectangular pulse; amplitude: 4.5-5.0 V; Duration: 8 microseconds; Timing resolution: less than 100 ns.



FIGURE 4. Dimensions of spectrometry CdZnTe detector Kromek.

5. Experimental Design and Set-up

The design of the measurement was based in the rate of gamma emission from the source of 60 Co with 500,000 Ci of radioactivity, the rate of photons emitted is 3.7×10^{16} gammas/s (1.17 and 1.33 MeV with practically yield = 100%). Note: The source of the irradiator on which the tests were performed has an activity of 500,000 Ci. The system was calibrated in energy with a window from 60 keV to 2 MeV. A first attempt was made to measure the source by removing it from the pool, placing the shielded detector as far as possible within the installation which was 4 m. The result was that the



FIGURE 5. Saturated gamma spectrum due to the high intensity of ⁶⁰Co gamma rays. The saturation is reflected as a continuous curve where the photopeaks are not visible.



FIGURE 6. Detector shielding schematic.

system became saturated, reflecting in a continuous spectrum where no photopeaks of the 60 C o energies were visible (Fig. 5).

It was considered to carry out the measurement by placing the shielded detector inside the pool, in addition. A container made of acrylic was made to protect the detector and introduce it into the water.

The detector shielding in the form of parallelepiped was constructed to have a circular collimation of 0.5 cm in diameter for the entrance of the gamma beam (Fig. 6). The rate of photons T_{pdD} detected at a distance D, without considering attenuation due to water and considering the collimation area (0.5 cm diameter) is:

$$T_{pdD}(c/s) = \frac{T_{\rm pe}}{S_D} A_c E f_{\rm in^{60}Co},\tag{1}$$

where: $E f_{in^{60}Co}$ is the intrinsic efficiency for ⁶⁰Co; T_{pe} represents the rate of photons emitted by the source, S_D is the area of sphere at distance D and A_c is the area of collimation. The photopeak efficiency was obtained for ⁶⁰Co (1.33 MeV) and it was 1×10^{-4} .

In Table I, shown the rate of photons through the collimated area of the detection system at distance D from the source (middle), attenuated by water; then, the counts registered (net count) in the photopeak of 1332.5 KeV in counting time of 900 s and the corresponding counting error.

TABLA I. Counts recorded in Photopeak of 1.3 MeV in 900 s of counting time.

| Col(A) | Col(B) | Col(C) | Col(D) |
|--------|----------|----------|--------|
| 2.0 | 1.63E+06 | 1.47E+05 | 0.3% |
| 2.5 | 8.62E+04 | 7.76E+03 | 1.1% |
| 3.0 | 1.66E+03 | 1.49E+02 | 8.2% |
| 4.0 | 2.48E+00 | 2.23E-01 | 212% |
| 5.0 | 9.47E-02 | 8.52E-03 | 1083% |

Col(A) Distance Source-Detector System D (m). Col(B) Rate of photons/Detector area * attenuated by water (s⁻¹), Col(C) Counts recorded in the photopeak (900 s). Col(D) Counting Error (%).



FIGURE 7. Experimental setup for ⁶⁰Co source measurement. The detector system was placed at a depth of 2.5 m in the gamma irradiator pool.



FIGURE 8. Handling the detector system in the installation.

A distance D = 2.5 m was chosen to perform the measurement, considering that a count rate of 857 c/s is a low enough not to saturate the system, and not so low to perform a counting of a long time. In Fig. 7 the array of the measuring is shown. In Figs. 8 and 9 the maneuver in pool is shown.

It is important to mention that the exposure speed for the half million Ci source at a distance of 5 m inside the pool is less than 2 μ Roentgen/h, so it does not represent a radiological risk for the personnel, however all the personnel who



FIGURE 9. Introducing the detector system inside the gamma irradiator pool, by means of a system of cables and pulleys.

participated in the manoeuvre are occupationally exposed to radiation (OEP), controlled through the licenses of use and possession of radioactive material granted by the National Commission of Nuclear Safety.

6. Results

The spectrum resulting from aforementioned measurements (integrated counting in 900s) shown the two photopeaks within the 1173.3 and 1332.5 keV centroids and a positive



FIGURE 10. Collected spectrum in which the photopeaks corresponding to the 60 Co gamma energies (1.17 and 1.33 MeV) are clearly shown.

correlation with the comptom effect continues component, which were found within the 0.5 keV uncertainty needed to conclude a positive identification corresponding to the energies from 60 Co, indicating strong evidence of the presence of the radionuclide (Fig. 10).

7. Discussion

The final obtained spectrum does not showed the pileup phenomenon as theorized when considering the number of events per unit time as planned, which led to the formation of the two ⁶⁰Co characteristic photopeaks. Moreover, their interaction energy with the detector strongly correlated with the FWHM by exhibiting values of 20.1 y 22.0 keV, which agree with the expected values of 20.2 y 21.9 ± 3 in accordance with the energy calibration.

Although the probability of interaction of gamma radiation in the detector by photoelectric effect is higher at lower energies, the 1.33 MeV photopeak appeared larger than that of 1.17 MeV, which could be explained by the energy attenuation caused by the path interaction with the pool water. This phenomenon has been observed before, and it is caused by the increased probability of interaction of low energy photons within the measurement medium. According to the measurements performed, the implemented method proved that it can be applied under measurement conditions with extremely high gamma emission intensities. One proposed important application is to verify the activity of ¹³⁷Cs in degraded fuel rods within power plants, which correlates with the degree of utilization of U as fuel to maximize plant efficiency, procedure that nowadays is generally approximated via numerical methods and lack of experimental verification, among others.

8. Conclusions

The method developed for the measurement of extremely intense sources for their identification of the radionuclide of interest ⁶⁰Co was successful. The use of a low efficiency and intermediate resolution CdZnTe detector, as well as the measurement in an attenuated medium was adequate to carry out the positive identification of ⁶⁰Co. Conditions established for the measurement resulted in a counting rate that did not saturate the system and allowed the spectrum to be collected to clearly appreciate the photopeaks in only 15 minutes of counting.

- G.F. Knoll, Radiation Detection and Measurement, third ed. John Wiley and Sons, New Jersey, 2000.
- G. Lutz, Semiconductors Radiation Detectors, Springer, Munich, Germany 1999.
- Kromek, Gr-05 User Manual, Rev 3, https://www. kromek.com/ U.K. 2022.
- Ortec, https://www.ortec-online.com/ electronics/ctr/, USA. 2022.
- IAEA, Livechart of Nuclides, https://www.nds.iaea. org/relnsd/vcharthtml/VChartHTML.html.

- S. Usman, A. Patil, Radiation detection deadtime and pile up: A review of the status of science. *Nuclear engineering and Technology*, **50** (2018), 1006.
- Sukhpal S. gamma-ray summing in germanium detectors and its effects on nuclear decay parameters. Asian Journal of Chemistry, Nov-2010.
- Ministerio de Defensa, Detección e identificación de material nuclear y radiológico. Estado de Arte y tendencia futura, España, 2009.
- Cember, Herman, Introduction to Health Physics, third edition, New York: McGraw-Hill, Health Professions Division, 1996.