

Study of Neutron Background in order to Improve Radioactive Waste Drum Characterization

Gabrielle Lelaizant¹
¹CEA, France
gabrielle.lelaizant@cea.fr

Abstract— A usual way of radioactive waste drums characterization combines gamma spectrometry measurements with passive neutron measurements. The CEA facility for waste drum characterization hereby described has been operated for more than 30 years. In this framework, a large variety of waste drums has been characterized in terms of spectra, densities, materials and radioactivity levels. As the facility was first dedicated to measure Intermediate-Level Long-lived Waste, the neutron background was not significant compared to expected neutron emitters from waste packages. These last years, Dismantling and Decommissioning operations have been advanced in this CEA site to the point where they are now associated with mostly Very Low Level Waste production. Therefore, neutron background is becoming significant. Using the large variety of past characterized drums brings the opportunity to study this background. The present study has been led over a sample of almost 1500 drums over a wide waste density range. These drums have been selected over the last 20 years by taking into account only one criterion: without any expected neutron emitters from the waste itself. This work first presents the technical settings of the measurement facility before describing the raw data of the measurements. Next, a statistical study over raw data enables to better acknowledge the neutron spallation background behavior in terms of time, density and materials. Ensues a way of using this new knowledge in order to improve how to take into account neutron spallation background in passive neutron measurements of packages of low actinides activities and high densities.

Keywords —neutron background, coincidence counting, radioactive waste characterization, cosmic-ray induced neutrons

I. INTRODUCTION

The combination of gamma spectrometry with passive neutron measurement has so far been a usual way to characterize radioactive waste [1]. Neutrons are identified and quantified using coincidence counting following thermalization in an assay chamber. The first device enables to quantify radioactivity in a waste package spectrum through the identification of its gamma emitters, whereas the latter not only

allows the measurement of nuclear material mass through the neutron count rate associated with spontaneous fission of fissile emitters (e.g. ^{240}Pu , the usual neutron tracer for plutonium assay) but also to confirm the measurement data obtained by gamma spectrometry. These combined measurement devices are well suited in case of alpha spectra, for which both measurements provide different pieces of information and then enable to measure actinides for a large range of waste densities. But some difficulties are encountered when alpha radionuclides activities are so low that they cannot be measured in reasonable measurement time durations in gamma spectrometry. In high density waste which prevents gamma signal from reaching detector cells, characterization with a high confidence level becomes a major issue.

Hence, the role of passive neutron measurement becomes even more important as it is not sensitive to the same parameters. On the contrary to gamma spectrometry, neutron absorption increases with the quantity of light matrices and especially with the proportion of low atomic number materials such as hydrogen. The fast neutrons emitted from spontaneous fission of the fertile isotopes provide good penetrability of bulk materials.

As the facility was first dedicated to measure Intermediate-Level Long-lived Waste (ILW-LL), gamma spectrometry has been paid more attention in waste characterization rather than neutron measurement results which are mainly used to get the value of nuclear mass. In this context, the neutron spallation background was not significant enough compared to expected neutron emitters from waste packages. Thus, the neutron background has been corrected so far by a fixed value which does not depend on the waste characteristics. In recent years, Dismantling and Decommissioning (D&D) operations have been advanced in this CEA site to the point that they are now mostly of Very Low Level Waste (VLLW) category. Therefore, considering a proper correction of neutron background level for each measurement is becoming a major issue in order to investigate low levels of neutron count rates from nuclear material.

Neutron background originates from different contributions [1] :

1. External-area radiation sources (e.g. waste storage area proximity);
2. Radioactive alpha or beta decay in the walls of the detector tubes;
3. (α , n) reactions in α emitters matrices;
4. External-area cosmic-ray sources (e.g. spallation reactions in the room);
5. Spallation reactions from cosmic rays in the sample or in the detector body.

Items 1 to 4 only produce single neutrons, whereas item 5 is accompanied by both single and coincidence neutrons.

Passive neutron coincidence counting is a well-known method for non-destructive assay of nuclear material contaminated waste [3] , [4] . Coincidence counting distinguishes the time-correlated neutrons, produced from fission, from neutrons born randomly in time such as those originating from items 1 to 4 (and mainly from #3's (α , n) reactions). Expressed as the equivalent mass of ^{240}Pu , the nuclear material detection limit is constrained by the cosmic-ray background of spallation neutron events taking place in the vicinity and body of the assay chamber, as well as in the waste itself. This background cannot be removed by a "simple" coincidence counting. Moreover, the cosmic-ray background of spallation neutron events increases with the volume and density of the He-3 tubes, as well as with waste density and is so matrix-dependent. Therefore, the waste itself becomes a spallation neutron source.

The intrinsic cosmic-ray background from the chamber can be minimized at the design stage by avoiding the use of materials of high atomic number in the detector vicinity. During operation, judicious location and shielding of the instrument can also help to reduce cosmic interaction in the chamber's body. Additional means of suppressing and stabilizing the cosmic-ray background are provided by data rejection algorithms, applied on an event-by-event basis by the software, to filter the major part of the cosmic-ray variability from the data, or to eliminate the high-multiplicity cosmic-ray neutron bursts. Nevertheless, no such algorithm can be applied in the current technique whose commissioning took place few decades ago. Besides, the electronics currently used do not allow the definition of a specific background for each measurement, but only a fixed value can be set as correction to calculate the net signal. This fixed value has been obtained from repeated measurements performed in the same configuration in a short period of time during the commissioning stage of the device. Also, any change of configuration leads to a new definition of this correction parameter. One may imagine a proper background correction depending on waste density. Indeed, some works have been carried out in order to define a material component as described in [1] and [5] . However, the results obtained in these studies cannot be considered as universal, but as equipment-dependent. The approach hereby presented deals with a data analysis study aiming at defining this specific correction on the basis of measurement feedback. As the end of

D&D operations is coming in this CEA site, the same electronics have been laid out in order to keep the same raw data format and shall remain so until the end of operations.

II. EXPERIMENTAL SETUP

A. Data selection criteria

Preliminary implicit criterion resides in no expected nuclear material. Some criteria have been taken into account over the whole dataset. First, the current data format has been applied only since 2008. Therefore, the last 11 years' data have been considered (2008–2019). From this dataset, only waste packages without any expected nuclear material (according to historical information provided by waste producers), were selected. 631 drums remain after this selection.

In order to avoid statistical effect on a single measurement, the waste packages (i.e. 200 L drums), are usually measured at least 3 times (and up to 10 times), with a duration of either 1800 seconds (the most common case) or 900 seconds (the former case). Each measurement result is considered in the dataset as an independent result. Nevertheless, a slight difference has to be specified here: the second applied criterion consisted in rejecting raw data which belong to a sample composed of the different measurements of the same drum but which show a standard deviation greater than 10% to the average value. 2050 measurement results remain after the application of the previous condition.

A comparison of each measurement value with its specific decision threshold enables to keep only significant raw data. 2.4% of the dataset was removed through this procedure. Finally, a statistical Grubbs test was applied to identify and discard outliers. 1797 measurement results compose the final dataset.

B. Passive Neutron Measurement Cavity and Variable Dead-Time System Electronics

The subsurface passive neutron measurement device is composed of 24 ^3He proportional counters (i.e. detector tubes) embedded in a HDPE layer. The 15 cm thick HDPE layer enables not only to minimize neutron background coming from the outside of the cavity but also to thermalize neutron energy in order to increase the cross-section of their capture into the ^3He tubes. Electronic modules are connected to the ^3He tubes in order to discriminate neutron events from γ pulses as well as to shape the output signal by summing up input signals from all electronics (one module for 3 ^3He tubes).

Passive neutron coincidence counting using shift register electronics is an established technique for non-destructive assay of nuclear material-contaminated waste. The "Variable Dead-Time System" (VDTS) electronics hereby chosen, following historical considerations, are slightly different. Even these modules are less common, they were well described in French standards [6] (in effect until 2020) as an alternative means to shift registers in order to address the neutron coincidence count rate.

As shown in Fig. 1, two counters are used in these electronics. The fast channel counter, named C_{fast} , increases by increments for each neutron event, whereas each first neutron event triggers the opening of a fixed time window named “dead-time τ_L ”. As the value of applied dead-time is equivalent to the detector die-away time, time-correlated neutrons from spontaneous fission are removed from the slow channel counting, C_{slow} .

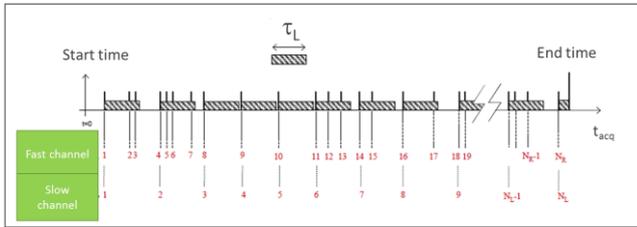


Fig. 1. Description of fast and slow channels' counters

Therefore, the difference between both counters C_i enables to calculate the number of correlated neutron events. Then, by applying the proper correction of times, i.e. the acquisition time t_{acq} and the dead-time τ_L , a coincident count rate X_i , equivalent to the spontaneous fission rate, is available when following the equation (1)

$$X_i = \left(\frac{C_{fast}}{t_{acq}} - \frac{C_i}{t_{acq} - C_i \cdot \tau_L} \right), \quad (1)$$

The second step consists in subtracting the background coincident count rate X_{bkg} in order to address the equivalent mass of ^{240}Pu directly related to the nuclear mass of the waste package. It is useful to remind here that any density correction is applied in these calculations.

C. Density characteristics of the dataset

However, the density of waste packages varies over a wide range: from 0.1 to 1.4. The histogram shown in Fig. 2 represents the density distribution of the 631 selected drums.

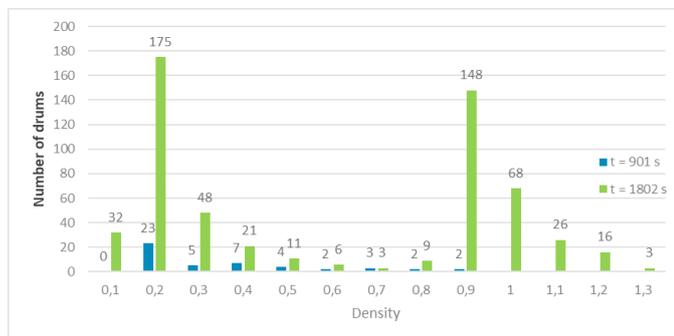


Fig. 2. Density distribution of selected drums

Two density ranges are mostly available in the selected dataset: 0.2 and ≥ 0.9 . These two density ranges are targeted in the following parts.

III. DATA ANALYSIS

A. Background coincident count rate versus density

As previously described, an increase of background coincident count rate is expected with density due to cosmic-ray-induced spallation neutron events. This effect is indeed highlighted in Fig. 3.

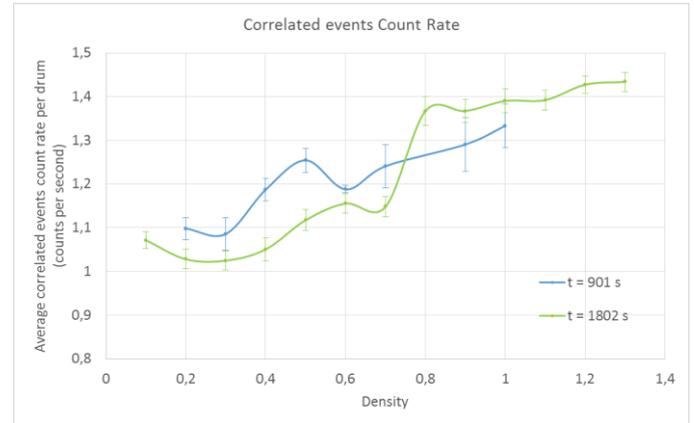


Fig. 3. Average correlated events count rate versus density for two different measurement durations (900 seconds in blue and 1800 seconds in green)

Error bars correspond to standard deviations (1σ) calculated over density samples for each density range. Even if the expected increase with density is visible in both plots, the green (1800 s) plot highlights a clearer cutoff at density ~ 0.8 as compared to the blue (900 s) plot. Smaller statistics available in shorter measurement durations (900 s vs 1800 s) dataset explains the observed flattening of the blue plot compared to the green one.

All background coincident count rates – without any averaging – are plotted in Fig. 4. The same behavior hereby appears in the shape of two ranges of background coincident count rates: one for light matrices (in the bottom part) and the other for heavy matrices (in the top part). Scatter plots in Fig. 4 express deviations to average.

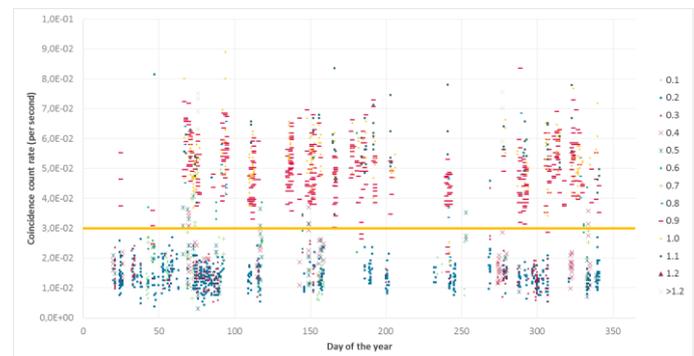


Fig. 4. Coincident count rates versus day of the year (between 0 and 365) for all densities showing two significantly different areas, according to matrix density (light to heavy)

TABLE I
 BACKGROUND COINCIDENT COUNT RATE PER DENSITY

| Density | Averaged background coincident count rate (counts per second) | Standard deviation |
|---------|---|--------------------|
| 0.2 | 0.014 | 0.002 |
| 0.9 | 0.051 | 0.011 |
| 1.0 | 0.053 | 0.007 |
| > 1.0 | 0.057 | 0.010 |

Table I highlights a 4-fold factor between high density and low density background coincident count rates.

Following the decision threshold as defined in equation (2),

$$DT = 2 \sqrt{\left(X_{\text{bkg}} + \left(\frac{C_{f,\text{bkg}}}{t_{\text{bkg}}} \right)^2 \cdot \tau_L \right) \cdot \left(\frac{1}{t_{\text{acq}}} + \frac{1}{t_{\text{bkg}}} \right)} \quad (2)$$

improving the background coincident count rate consideration enables to better calculate the decision threshold and consequently to better consider significant signals. The 4-fold factor previously highlighted in Table I also stems from this equation.

B. Background coincident count rate versus time

The neutron background arises primarily from cosmic origin with negligible contributions from radioisotopes in the local environment [7]. The cosmic-ray-induced neutrons observable at ground level are mainly produced through spallation reactions on nitrogen, oxygen and argon nuclei in air from high-energy particles (mainly protons and helium ions).

Neutrons can originate from miscellaneous sources either diffuse or point-like [8]:

- Solar flares (neutrons, either from direct emission from the Sun or as secondary products in the Earth's atmosphere);
- Galactic neutrons (relativistic neutrons created in active galactic nuclei);
- Cosmic neutrons (from supernova remnants or other high energy cosmic ray point sources);
- Atmospheric neutrons (e.g. secondary neutrons produced by cosmic-ray induced hadronic showers in the Earth atmosphere).

According to [8], solar neutrons are the most likely source of spallation neutrons.

Therefore, a time-dependent background coincident count rate may be inferred following either solar flares or different solar cycles, ranging from the shortest (day/night cycle), to the longest period (eleven years from the heliospheric and geomagnetic fields modulations [9]). The impact of solar wind could also be investigated as a cosmic ray flux has been shown to be inversely correlated with the Sun's activity-generated solar winds [7].

Fig. 5 plots the background coincident count rate versus the day of the year since 2008, for the four density ranges introduced in Fig. 4. Both yearly effect and seasonal effect

were targeted in these plots. Horizontal lines correspond to respective averaged background coincident count rates.

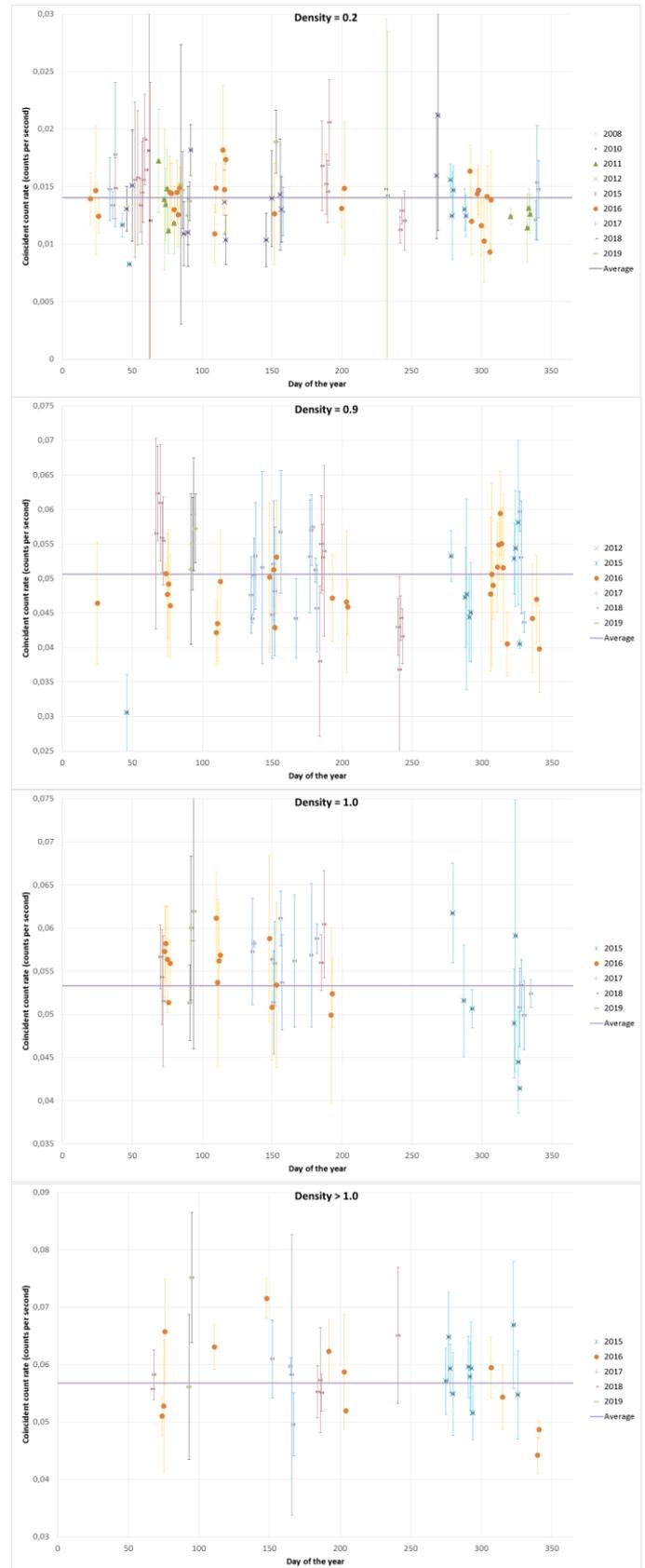


Fig. 5. Coincident count rates versus day of the year

Error bars correspond to standard deviations (1σ) over the sample for each day of the year. By considering error bars as uncertainties over the dataset, a greater deviation to the average value could be characteristic of higher densities. In this case, more discrepancies (i.e. isolated points) could show time effects related to cosmic rays events or solar cycles. Nevertheless, establishing a clear relation is complex, owing to different time scales between measurement times (hundreds of seconds) and solar flares durations (minutes, less than 100 seconds) or cosmic events durations (seconds, up to minutes for the most energetic phenomena).

When searching for an hourly effect during the day in Fig. 6, deviations to average are even less significant.

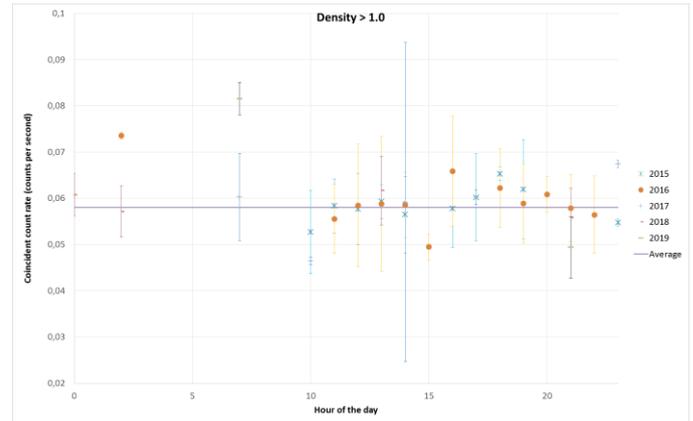
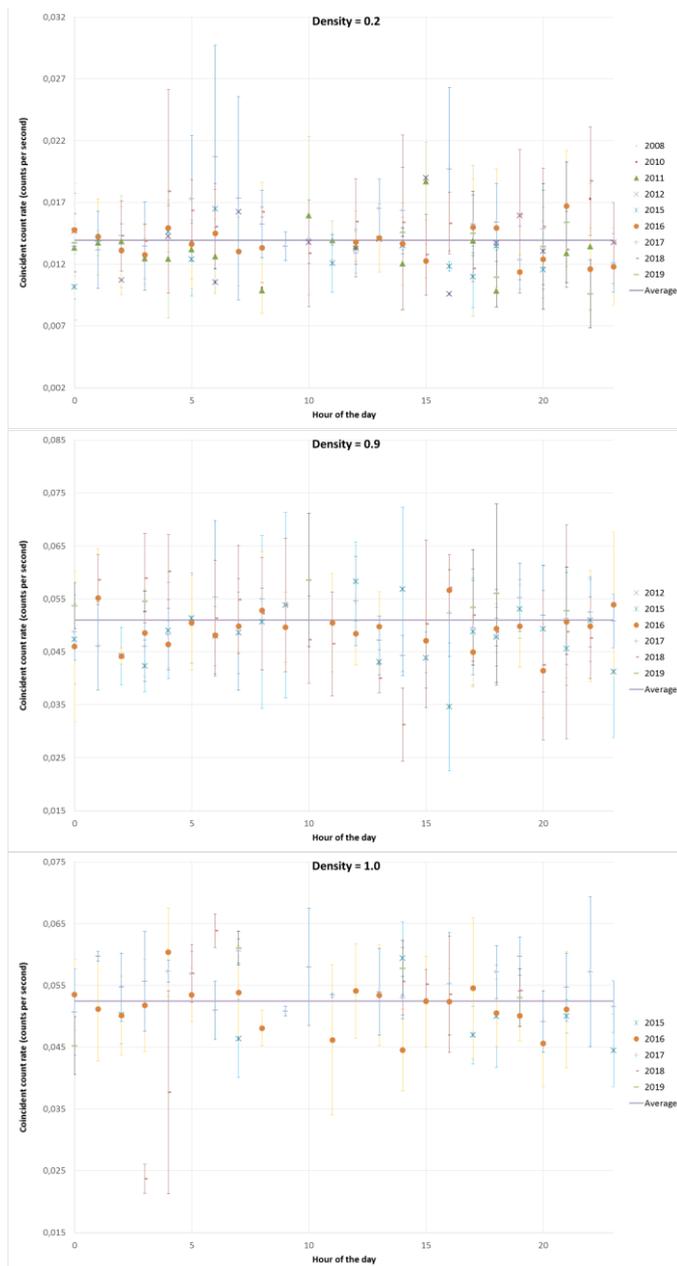


Fig. 6. Coincident count rates versus hour of the day

IV. CONCLUSION

The data analysis study presented here over the analysis of 2050 measurements undertaken over 11 years has confirmed the expected density effect on background coincident count rate observed in passive neutron measurements, due to stronger spallation effects in higher densities materials. Density-dependent values have been defined from the feedback study in order to improve background consideration not only to better calculate net signal by applying a proper background correction, but also to take into account the right decision threshold in order to discard insignificant measurement data.

The search for time-dependent variations related to galactic or extragalactic cosmic rays events or solar cycles has not been fruitful but could be improved by using a timestamp module in order to investigate neutron flares in shorter time windows.

REFERENCES

- [1] IAEA, Strategy and Methodology for Radioactive Waste Characterization, IAEA-TECDOC-1537, Vienna, Austria, 2007, pp. 66-72
- [2] H.O. Menlove, D.H. Beddingfield and M.M. Pickrell, The Design of a High-efficiency Neutron Counter for Waste Drums to Provide Optimized Sensitivity for Plutonium Assay, https://www.canberra.com/fr/literature/waste_special_systems/tech_papers/menlove.pdf
- [3] G.F. Knoll, Radiation Detection and Measurement, New York, John Wiley and Sons, 1999
- [4] D. Reilly, N. Ensslin and H. Smith Jr, Passive Nondestructive Assay of Nuclear Materials, NuREG/CR-5550,LA-UR-90-732, 1991, pp. 457-528
- [5] S. Croft and L.C.A. Bourva, The specific total and coincidence cosmic-ray-induced neutron production rates in materials, Nuclear Inst. And Methods in Physics Research A 505 (2003) 536-539
- [6] NF M60-306:1998 Standard, Nuclear energy – Fuel cycle technology – Waste – Determination of radioactive characteristics of solid waste packages by passive neutron counting
- [7] M.F. Becchetti, M. Flaska, S.D. Clarke, S.A. Pozzi, Measurements and simulations of the cosmic-ray-induced neutron background, Nuclear Inst. And Methods in Physics Research A 777 (2015) 1-5
- [8] D. Casadei, Neutron Astronomy, School of Physics and Astronomy, University of Birmingham and School of Engineering, FHNW, arXiv:astro-ph/1701.02788, 2017
- [9] S.E. Forbush, Cosmic-ray intensity variations during two solar cycles, Journal of Geophysical Research Vol. 63, 1958, pp. 651-669