

# **COMPACT NOVEL GAMMA / NEUTRON SIMULTANEOUS NUCLIDE IDENTIFICATION SYSTEM, FOR REAL-TIME VERIFICATION OF SNM, REMOTE INSPECTIONS OF UNATTENDED CYLINDER AND SPENT FUEL CASKS**

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## **Abstract**

A new light and compact NDA measuring system, allows operators to execute gamma and neutron passive NDA measurements and is capable to identify SNM through gamma and neutron identification simultaneously. Moreover, the identification of neutron sources such as Cf-252, Am-Be, Pu, U, Am-Li, Pu-Be, etc. is possible also in presence of high gamma field, and covers scenarios where is present heavy shielding, moderating material, and masking sources. The presented NDA instrument, weights 7.5 kg, including battery guaranteeing > 8 h continuous operation. It uses two detectors, runs Pulse Shape Discrimination in real time and includes an intelligent processing algorithm (patent pending) that combines multiple parameters of gamma and neutron radiation to uniquely characterize the material under essay. The basis of this technology is already in use in the fast neutron collar deployed by the IAEA Safeguards for fresh fuel verification (FNCL system), and the product here presented, called SNIPER-GN, is a variation conceived as unattended verification station both for cylinders as well as for casks.

The paper is describing the results of SNM identification and measurements in different configurations and is extrapolating and describing the potential use in all major identified unattended scenarios. The system could be used also in attended mode and is already design for scalability to variate detection efficiency as well as different level of gamma resolution. The system is capable to determine in passive mode ranges of enrichment level both for Pu-239 and U-235.

The system could offer an opportunity to verify the possibility in the future fissile material measurements through the cask wall verification of shielded/unshielded casks, NDA for partial defect testing and with some additional R&D could be enhanced with position sensitive integrated technologies such as fast segmented plastic detectors with fast DAQ processing for multi-assembly configurations, defect tests of spent fuel assemblies.

## **1. INTRODUCTION**

This paper presents the new features of a portable radioactive isotope identifier for the detection and identification of both gamma and neutron emitting radionuclides, even multiple ones. This new technology was already introduced the first time in paper [1]. The new features introduced can be resumed in:

- the capability to identify sources through the detection of neutrons, discriminating spontaneous fission sources (Cf-252),  $\alpha$ -n sources (Am/Be, Am/Li) and nuclear material containing mix of isotopes of plutonium or uranium (extensive tests results)
- the capability to make cross correlation between gamma and neutron measurements to achieve a higher level of accuracy in the identification of SNM that emits both neutron and characteristic gamma lines

Many measurements have been performed in several accredited laboratories with different mix of SNM sources.

Different measurement conditions were tested at gradually increasing levels of difficulty, adding, for example: shields, moderators, masking gamma sources and a mix of the above mentioned. Before reporting the tests setup with their results there is an introduction on the instrument technology and the implemented algorithm.

## 2. PSD ALGORITHM AND SNM IDENTIFICATION

The device used for the tests presented in this paper is equipped with an organic liquid scintillator with excellent Pulse Shape Discrimination (PSD) proprieties for the simultaneous detection of gamma rays and neutrons and a mid-high resolution inorganic crystal for gamma spectroscopy identification. The system here presented makes use of a CAEN DT5790, an advanced digital Multi Channel Analyzer (MCA) running a Digital Pulse Processing (DPP) firmware in the FPGA. The PSD firmware performs online fast discrimination of neutrons from gammas, enabling individual alarms for each kind of particle. The PSD algorithms based on a digital dual gate charge integration technique able to sustain high counting rates. It performs double integration of both prompt and total charge, self-triggering, input signal baseline calculation and pedestal subtraction for energy calculation and pile-up rejection. The anode signals of both detectors are fed directly into the CAEN DT5790 digitizer (2 x 250 MS/s 12-bit). The DPP-PSD algorithm provides on-line data for each event like: (a) the time stamp, (b) the total charge integration of the signal, (c) the partial charge integration of the signal used for PSD and (d) the possibility of storing a selected part of the digitized signal. The PSD capabilities of the detector are so exploited in real-time to discriminate gamma from neutrons. The same algorithm has been designed and implemented by CAEN and is today in use at IAEA safeguard department in the Fast Neutron Collar Monitor for fresh fuel verification (as described in [2]). This device can detect radioactive source as Special Nuclear Material (SNM), medical, industrial and Naturally Occurring Radioactive Material (NORM). The main ability of this instrument is not limited to counting neutrons, but it can also identify the neutron sources. In addition, it can discriminate between different fission sources (Cf-252)  $\alpha$ -n sources

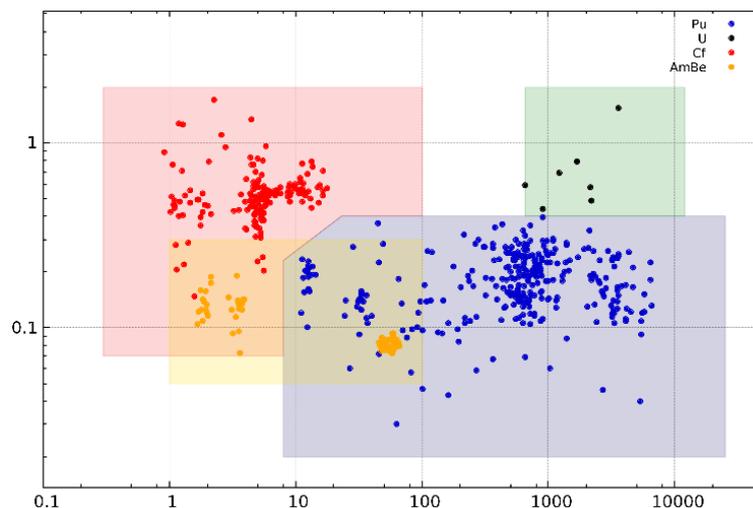


FIG. 1. Neutron source identification algorithm results. Each dot represents a 1-minute acquisition for the different sources in different condition: naked source, poly shielded, lead shielded, mixed shielded or gamma masked. The overlap of the Am-Be sources is resolved with a third parameter of the algorithm.

(Am/Be, Am/Li) and nuclear material containing mix of isotopes of plutonium or uranium (see FIG. 1). The identification algorithm performs a subtraction of environmental background spectrum from the one-minute spectrum accumulated during the identification measurement.

Then, multiplicity rate and analysis of energy spectrum of neutrons are performed on the obtained spectrum.

The measurements characteristics were already introduced in the [1] and they are listed here for convenience:

- A moving average is used to integrate the last three minutes of natural background in order to set individual thresholds for neutron and gamma counts;
- Alarms are triggered separately when the respective rate exceed the alarm thresholds. These thresholds are calculated to allow detection with 95% detection probability at 95% confidence level for a dose rate on the front face of the scintillator of at least 50 nSv/h;
- The neutron source detection has been tested also in high gamma ray fields up to 0.1 mSv/h.
- The novel discrimination algorithm allows also a neutron source classification
- The measured FoM (see caption of Fig. 2) with an energy window of  $480 \pm 75$  keV is

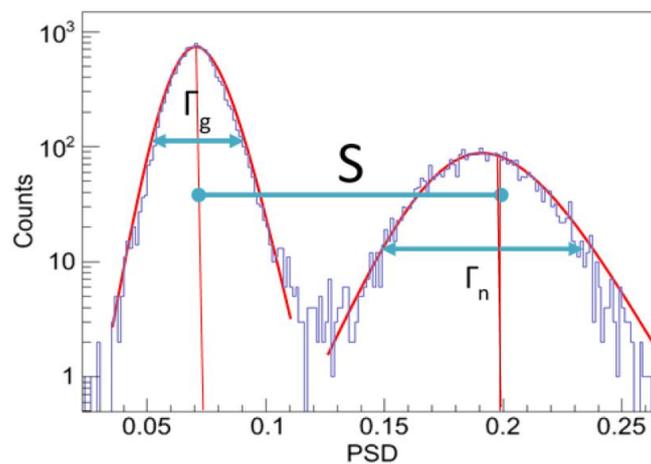


FIG. 2. FoM plot obtained by projecting the PSD plot on the x axis. FWHMs and separation between centroids are shown in the plot.

1.73. the FoM “Figure of Merit” indicates the performance of the Pulse Shape Discrimination. The graph of the FoM is a projection of the y axis of the PSD plot on the x axis thus obtaining a PSD vs count plot with two Gaussian distribution, one for the gamma and one for the neutron.

The FoM is defined as:

$$FOM = S / (\Gamma_n + \Gamma_g)$$

where S is the distance between the two centroids of neutron and gamma gaussian distribution and  $(\Gamma_n + \Gamma_g)$  is the sum of gamma and neutron FWHMs.

### 3. MEASUREMENT CAMPAIGN

Many different tests were performed with the device in the new compact configuration. Most of the measurements were focused on the neutron source identification (see Figure 1). The identification measurement was performed using a one-minute acquisition. This is the maximum time allowed by the standard.

#### 3.1. Test Session 1

Two different sources were available at the laboratory and were used in this session. Each source was composed by a mixture of Plutonium isotopes (such as Pu-242, Pu-241, Pu-240, Pu-239, Pu-238) and their decay products. The two sources differed in their enrichment level in Pu-239. These sources were encapsulated in a cylindrical shell of 0.6 cm stainless steel. For security purposes CAEN SpA is not allowed to share more information on the source composition and amounts. The neutron yields at the measurement date were:

TABLE 1. Neutron emission rate at session 1

Source name	Neutrons/s emission at the test date
Source-1	4,169
Source-2	3,065

Concerning the neutron alarm there are not any specification on the neutron emission for the Pu in the standard. For this reason the same parameters used for the Cf-252 were kept.

Moreover, There are not specifications or limitation on the parameters for the identification performed through neutron detection because there is not any instrument that can perform this type of measurement yet. For the neutron static measurement the standard reference of a Cf-252 source emitting 20.000 n/s placed at 25 cm has been used. The standard distance (25 cm) has been rescaled proportionally to the neutron emission rate of the Plutonium source. Considering a neutron emission that cover the full available solid angle ( $4\pi$ ) the proportion used is:

$$\frac{20.000 \left[\frac{n}{s}\right]}{4\pi * (25 [cm])^2} = \frac{n \text{ emission source}_i \left[\frac{n}{s}\right]}{4\pi * (distance_i^{rif} [cm])^2} \quad (2)$$

Where the  $n \text{ emission source}_i$  are the emission rates listed in the previous table and the  $distance_i^{rif}$  are the new reference distances scaled on the used sources. The experimental apparatus has been placed in the different laboratories following the indications listed below:

- The device and source were placed on a support at least 50 cm above the ground to reduce the environmental radioactivity contribution to the background and avoid soil reflections of neutrons.
- The space between sources and device was empty to reduce the reflection of neutrons.
- The space around the measurement region was empty, to reduce the reflection of neutrons.
- The device/source distance for the first measurement was the one calculated with formula (1). If the identification result was correct the following steps were followed:
  - Increase the device/source distance by keeping the same shielding.
  - Increase the shielding of the source and the device by keeping the same source distance.
  - Increase both.

In the following tables the results of the identification through neutron detection are reported. There is not a standard or any requirement on the identification of source through the neutron detection because there is not any instrument that can perform this kind of measurements. The gamma spectrum identification result is not reported to not confuse the reader. It is worth noting that gamma measurement performance has been tested in another measurement session and the device exceeds the standard performance. The results of gamma identification in this case would have only double confirmed the results of the neutron identification measurement. In some of the reported measurements the neutron identification algorithm was forced to start manually because the source/detector distance was too high to trigger the neutron alarm (statistic too low). This fact shows that the neutron identification

algorithm is so performant that it can identify the neutron source also when the source/detector distance is so big that the counting statistic is not enough to trigger the alarm.

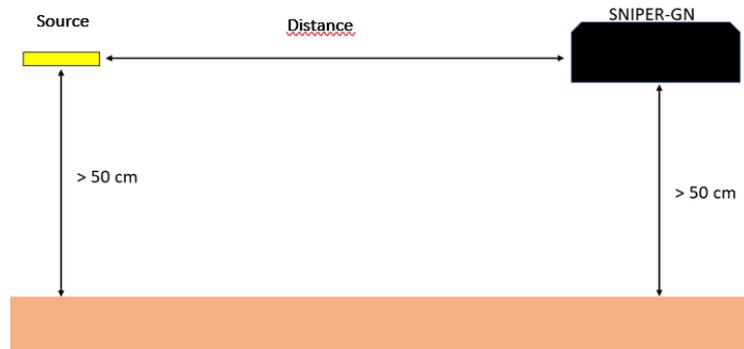


FIG. 3. FoM plot obtained by projecting the PSD plot on the x axis. FWHMs and separation between centroids are shown in the plot.

### 3.1.1. Source 1

For source 1 the  $distance_i^{rif}$  was 10 cm.

TABLE 2. Test results for the source 1 in different measurement conditions

distance (cm)	Shielding	Identification/trials
15 x standard	-	150/150
15 x standard	Poly 5 cm	14/15
15 x standard	Pb 5 cm	14/15
25 x standard	-	30/30

The maximum distance reached was 250 cm. It was 25 times greater than the standard distance.

### 3.1.2. Source 2

For source 2 the  $distance_i^{rif}$  was 10 cm.

TABLE 3. Test results for the source 2 in different measurement conditions

distance (cm)	Shielding	Identification/trials
15 x standard	-	9/9
15 x standard	Poly 5 cm	8/8
15 x standard	Pb 5 cm	6/8
20 x standard	-	8/8
25 x standard	-	12/15

The maximum distance reached was 250 cm with a success rate of 80% due to the lower neutron flux respect to source 1. It was 25 times greater than the standard distance.

## 3.2. Test Session 2

The source available at the lab were a Cf-252 source and a Am/Be source. The neutron yields at the measurement date were:

TABLE 4. Neutron emission rate at session 2

Source name	Neutron/s emission at the test date
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Cf-252	65,69
AmBe	198,75

The same considerations of the session 1 were applied here. In the following subsection the results of the identification through neutron detection are reported.

### 3.2.3. Cf-252

For the Cf-252 the  $distance_i^{rif}$  was 45.5 cm.

TABLE 5. Test results for the Cf-252 source in different measurement conditions

Distance (cm)	Shielding	Success/trials
Standard	Pb 5 cm + Poly 6 cm	10/10
2 x standard	Poly 10 cm	10/10
4.5 x standard	-	15/15

The maximum distance reached was 200 cm. It was 4.5 times greater than the standard distance.

### 3.2.4. Am/Be

For the Am/Be the  $distance_i^{rif}$  was 80 cm.

TABLE 6. Test results for the Am/Be source in different measurement conditions

distance (cm)	Shielding	Success/trials
standard	-	10/10
standard	Pb 5 cm + Poly 6 cm	15/15
2 x standard	-	15/15

The maximum distance reached was 160 cm. It was 2 times greater than the standard distance.

## 3.3. Test Session 3

In this measurement session the same sources of the session 1 were used. In addition, for some measurements, a Cs-137 source with an activity of  $4.5 \times 10^5$  Bq was added for masking purposes. The goals of this new session of measurements were:

- increase the difficulty of the measurement respect to session 1 by adding masking source or masking source plus shielding;
- test the first version of the cross-correlation algorithm that allows to double check the presence of SNM through both the neutron and the gamma measurements.

In the tables below the results of the identification measurement are reported. In the report column the nuclides coming from the neutron identification algorithm are reported with a \*, the nuclides coming from the gamma peak search algorithm performed on the mid-high resolution scintillator spectra are reported normally (without the \*).



FIG. 4. Pictures of the experimental setup of the Test Session 3

### 3.3.5. Source 1

For source 1 the  $distance_i^{rif}$  was 10 cm.

TABLE 7. Test results for the source 1 in different measurement conditions

N of trials	distance [cm]	Shielding [cm]	Report	Triggered alarm
10	10	-	Pu* Pu 100% CL	n and $\gamma$
15	10	5 Lead	Pu* Pu 100% CL	n and $\gamma$
17	10	5 Lead + 5 Poly	Pu* Pu 90% CL	n and $\gamma$
10	40	-	Pu* Pu 100% CL	n and $\gamma$
10	40	5 Lead	Pu* Pu 80% CL	n and $\gamma$
10	40	5 Lead + 5 Poly	Pu* Pu 80% CL	n and $\gamma$

### 3.3.6. Source 2

For source 2 the  $distance_i^{rif}$  was 10 cm.

TABLE 8. Test results for the source 2 in different measurement conditions

N of trials	distance (cm)	Shielding [cm]/Exposure from Cs-137 [nSv/h]	Report	Triggered alarm
11	50	- 0	Pu* Pu 100% CL	n and $\gamma$
5	25	- 452	Pu* Pu 90% CL Cs-137 100% CL	n and $\gamma$
5	25	5 Lead -	Pu*	n
5	25	5 Poly 473	Pu* Cs-137 100% CL Pu 90% CL	n and $\gamma$

5	10	5 Poly -	Pu*	n
5	10	5 Lead + 5 Poly -	Pu*	n
5	10	5 Lead 2963	Pu* Cs-137 100% CL Pu 100% CL	n and $\gamma$

From the above tables some considerations can be done:

- Respect to the measurement of the session 1 and 2 the gamma identification results were reported. In these reports the user can double check the confirmation of the Plutonium presence:
  - the one without the \* is obtained through the Pu emission lines identification in the mid-high resolution scintillator spectrum. The result is here accompanied with the Confidence Level value expressed in %
  - the one with the \* comes from the neutron identification algorithm (like in measurement of section 1 and 2) and the Confidence Level definition is currently under development.

#### 4. CONCEPTUAL DESIGN FOR UNATTENDED MEASUREMENTS

The above-described portable radioactive isotope identifier can be used as a Non-Destructive Assay device for inspection and can be also integrated in unattended monitoring stations.

The identifier integrates an ARM based CPU, which gives the possibility to schedule tasks, while running continuous data acquisition, and communicate through secured TCP/IP protocols. This allows the Safeguards unattended monitoring to have a continuous radiation level measurement through gamma and neutron counting, data saving and remote synchronization. Counts can be saved in configurable time windows and automatic identifications of radionuclides can be performed in parallel, to trigger alarms on the radiation level and on the type of radionuclide identified. For such features, the portable radioactive isotope identifier could be to provide a set of measured quantities that could effectively fit with the use in fuel processing facilities or spent fuel repositories.

An example of possible use of the isotope identifier is in the characterization of  $UF_6$  cylinders in nuclear processing facilities. The system can perform a measurement of the isotopic ratio with its mid-resolution  $CeBr_3$  inorganic scintillators and this information can be verified and related to the system characterization of the neutron-emitting radionuclides. The neutron emission provides information on the fissile nuclear material that is present deeper inside the volume of the cylinder, giving potentially complementary attributes to the  $UF_6$  cylinder. FIG. 5 represents a sketch of the possible layout of a  $UF_6$  cylinder verification. The portable radioactive isotope identifier could also be redesigned in a layout to connect external high-efficiency detector panels that can be arranged in the proximity of the  $UF_6$  cylinder, to reproduce the functionality of the FNCL in use by the IAEA, where neutron coincidence counting is performed with activation sources. This latter represents another complementary verification technique to the gamma spectrometry and the multiparametric analysis above described.

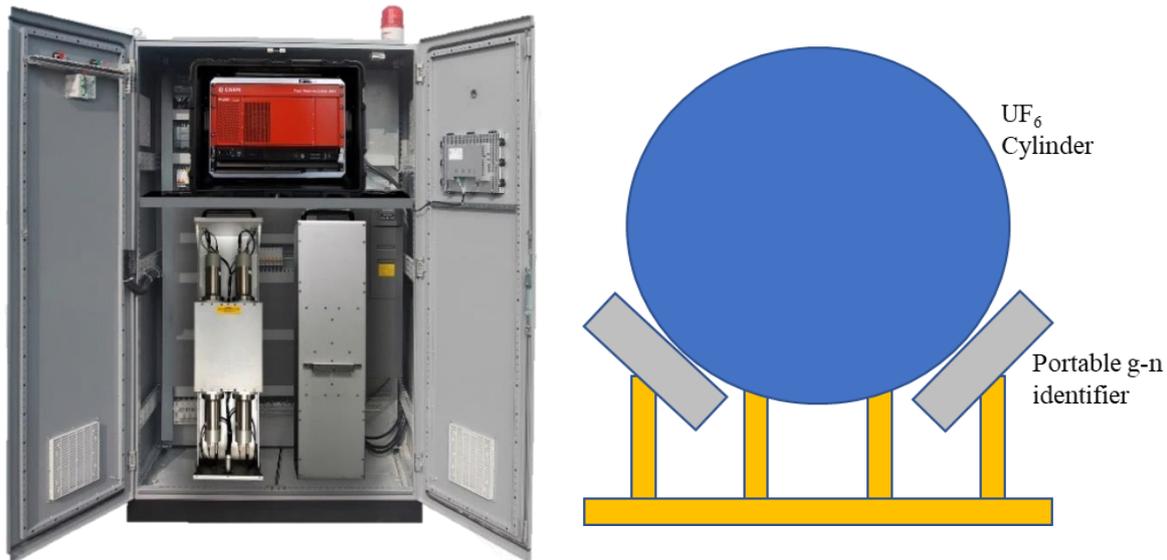


FIG. 5. On the left side the conceptual design of a monitoring station composed by high efficiency and segmented detector measurement system based on the portable radioactive isotope identifier. On the right side the conceptual design of the measurement setup per  $UF_6$  cylinders with the portable identifier displaced on its sides.

The unique capability of the system to detect plutonium could also be exploited to measure its presence or obtain observable quantities for the verification of spent fuel casks. The verification can be needed in case there is an event that has determined a loss of the continuity of knowledge on the identity and the origin of the spent fuel cask. The multivariate analysis performed on the external surface of the spent fuel cask could provide a footprint of the content to detect changes in its material arrangement. This technique could be paired with other experimental techniques as the muon tomography to detect the removal or the replacement of the content of the spent fuel cask.

## 5. CONCLUSIONS

The new patented algorithm applied to the neutron/gamma liquid scintillator spectrum allows to identify the neutron emitting sources such as Cf-252, Am/Be, U and Pu mixtures in different measurement conditions. This paper shows this capability extensively. The results reported in the tables from 2 to 7 clearly show the identification capability of the device. Moreover, the new powerful and compact electronics allows the use of two different type of detectors simultaneously while keeping a small form factor. The digital acquisition electronics also gives the capability to integrated segmented and high efficiency detection systems for unattended characterization of fuel cylinders in processing facilities and the verification of spent fuel casks in case of loss of continuity of knowledge. The double detector capability improves the identification performance of the gamma emitters. A high-resolution inorganic scintillator, such as  $CeBr_3$ , has been easily integrated into the system to perform an enhanced gamma radioisotope identification by characteristic gamma lines recognition. This solution allows for a simultaneous identification of multiple radionuclides, exceeding the limits of gamma identification performed with organic liquid scintillator and Compton spectra libraries only. The inorganic scintillator also enhances the neutron source identification capability for all radionuclides that are both gamma and neutron emitters, providing the end user with a double confirmation of the identification.

## REFERENCES

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