Acquisition of neutron-induced gamma signatures of chemical agents

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Abstract. The detection of special chemical elements that could be used by terrorist in improvised explosive devices (IED) can be performed by the spectroscopic analysis of neutron-induced gamma rays. Capture gamma-ray detection is optimally performed by thermal neutron analysis (TNA) between the pulses delivered by a fast neutron generator, when the ratio between thermal and fast neutrons reaches a maximum. On the other hand, gamma rays induced by fast neutrons, mainly during inelastic scattering reactions, can be optimally detected with the associated particle technique (APT). The signal associated to an inspected object is enhanced with respect to the noise coming from its environment through the selection of the gamma rays produced in the corresponding volume. In the frame of the French CBNR R&D program, which aims at developing new detection technologies to improve the fight against chemical, biological, nuclear, radiological and explosive threats, the combination of TNA and APT in a portable inspection system is being studied to identify the largest panel of chemical elements of interest, such as fluorine, sodium, phosphorus, sulphur, chlorine, arsenic, bromine, iodine, mercury and thallium. TNA and APT signatures of these elements are being acquired with low background measurement systems and long acquisition times, to constitute a reference database that will be used to process the data in a future mock-up of the portable inspection system. The status of the experimental database is presented and the measured gamma-ray spectra are compared with numerical simulations performed with the MCNPX particle transport code and the ENDF/B-VII nuclear data library.

1. Introduction

The French CBNR R&D program aims at developing new detection technologies and systems to improve the fight against chemical, biological, nuclear and radiological threats. In particular, neutron-induced gamma radiations can be used to detect chemical elements constituting an improvised chemical device possibly manufactured by terrorists. Portable systems based on a light transportable sealed tube neutron generator, which produces 14 MeV neutrons through the $^{3}H(^{2}H,n)^{4}He$ fusion reaction, and specific gamma-ray detectors, electronics and software, are being developed by industrials such as EADS SODERN [1]. This paper deals with an R&D program conducted by three CEA Divisions (Nuclear Energy, Military Applications and Technological Research) in view to specify the technical and operational needs, to propose and test innovative solutions, and to provide experimental data to support industrial developments in this field.
The work focuses on the detection of fluorine, sodium, phosphorus, sulphur, chlorine, arsenic, bromine, iodine and mercury. The detection of gamma rays produced by fast and thermal neutrons is studied to detect the largest number of elements. Capture gamma rays are recorded between pulses delivered by the electric neutron generator, when the signal-to-noise ratio (SNR) reaches a maximum. This is the classical acquisition method used in thermal neutron analysis (TNA) [2]. The gamma rays produced by the fast neutrons during the pulses can also be detected, but the associated particle technique (APT) [3], which allows a 3D separation of the suspicious object (e.g. a luggage abandoned in a public area) from its environment (wall, floor, etc.) and a subtraction of the background produced by reactions others than the ones induced by fast neutrons, leads to a better SNR and detection sensitivity. Each neutron produced by the $^{3}H(^{2}H,n)^{4}He$ reaction is emitted nearly back to back with an “associated”

alpha particle, which is detected by a position-sensitive detector embedded in the neutron generator. The precision on the neutron direction is related to the 2D resolution of the position-sensitive detector. On the other hand, the neutron time of flight is determined from the coincidence time measured between the detection of the alpha particle and the neutron-induced gamma ray. The 14 MeV neutron speed being about 5 cm/ns, a nanosecond time resolution is needed to reach a practical spatial resolution. Each neutron associated to the detection of an alpha particle is called a "tagged" neutron.

Parametrical studies by numerical simulation have been performed [4] to provide preliminary indications about the main features of the portable system: geometry, gamma-ray detectors, collimators, shields, neutron emission levels, useful signal, background noise, counting statistics. Experiments are being conducted with laboratory setups to verify calculation expectations and establish the practical detection capabilities of the system. Some of these tests are reported in [4] for APT, showing promising results for fluorine, sodium, phosphorus, sulphur and chlorine detection in a short acquisition time, with a light and compact system.

Experiments are also being conducted to acquire an extended library of APT and TNA gamma-ray spectra for all the elements of interest, which will be used for further data processing. These acquisitions are not performed with the portable setup but with large, heavily shielded, optimized systems, as well as long acquisition times to maximize precision. A comprehensive experimental database is required because significant lacks were already noticed in the available gamma-ray production data, leading to significant discrepancies between experiment and calculation [4,5]. Preliminary signatures are reported in [4] and this paper presents the last developments of the database.

Thallium, which was not available so far, has been investigated in the APT mode. On the other hand, due to a limited QDC range of the available electronics, APT spectra recorded so far extended approximately from 0.6 to 8 MeV. Indeed, the EURITRACK system used for these acquisitions is devoted to the detection of explosives and illicit drugs in cargo containers which, due to their size, induce large photon attenuation effects and do not allow for an efficient detection of low energy gamma rays. However, for the present work, several elements of interest did not show useable gamma rays in this energy area, such as arsenic, bromine, iodine and mercury [4]. Therefore, additional acquisitions have been performed with a larger PMT voltage to investigate the low energy region of the gamma-ray spectra, which now extend from about 0.1 to 1.6 MeV. However, due to self-absorption effects in the bulk samples used for these acquisitions, a physical cut is observed below 200 keV in the reported gamma ray spectra. Nevertheless, the low energy region can be exploited for the inspection of small objects with limited attenuation, the spectra presented in Section 2 showing useable signatures for a number of the above elements.

Concerning TNA signatures, experiments are underway with a PFTNA (Pulsed Fast and Thermal Neutron Analysis) installation available on CEA/DIF [7]. This measurement system allows to record inelastic and thermal spectra with a very good SNR. The decay spectra after irradiation were also recorded. The comparison of TNA (and APT) acquisitions with MCNPX simulations using the ENDF/B-VII database already showed discrepancies for sulphur [4]. The lacks in the gamma-ray production data are detailed here and new investigations with bromine are presented.

2. APT signatures

The APT gamma-ray spectra associated to the elements of interest have been recorded with six 5"×5"×10" NaI(Tl) detectors of the EURITRACK system and several hours of acquisition time. The setup and principle of these gamma signature acquisitions is detailed in [4].

The APT gamma-ray spectra associated to thallium, arsenic oxide, bromine, iodine and mercury are presented in *FIG.1*. Only the low-energy acquisitions are reported because no noticeable gamma ray were observed in the high energy region.

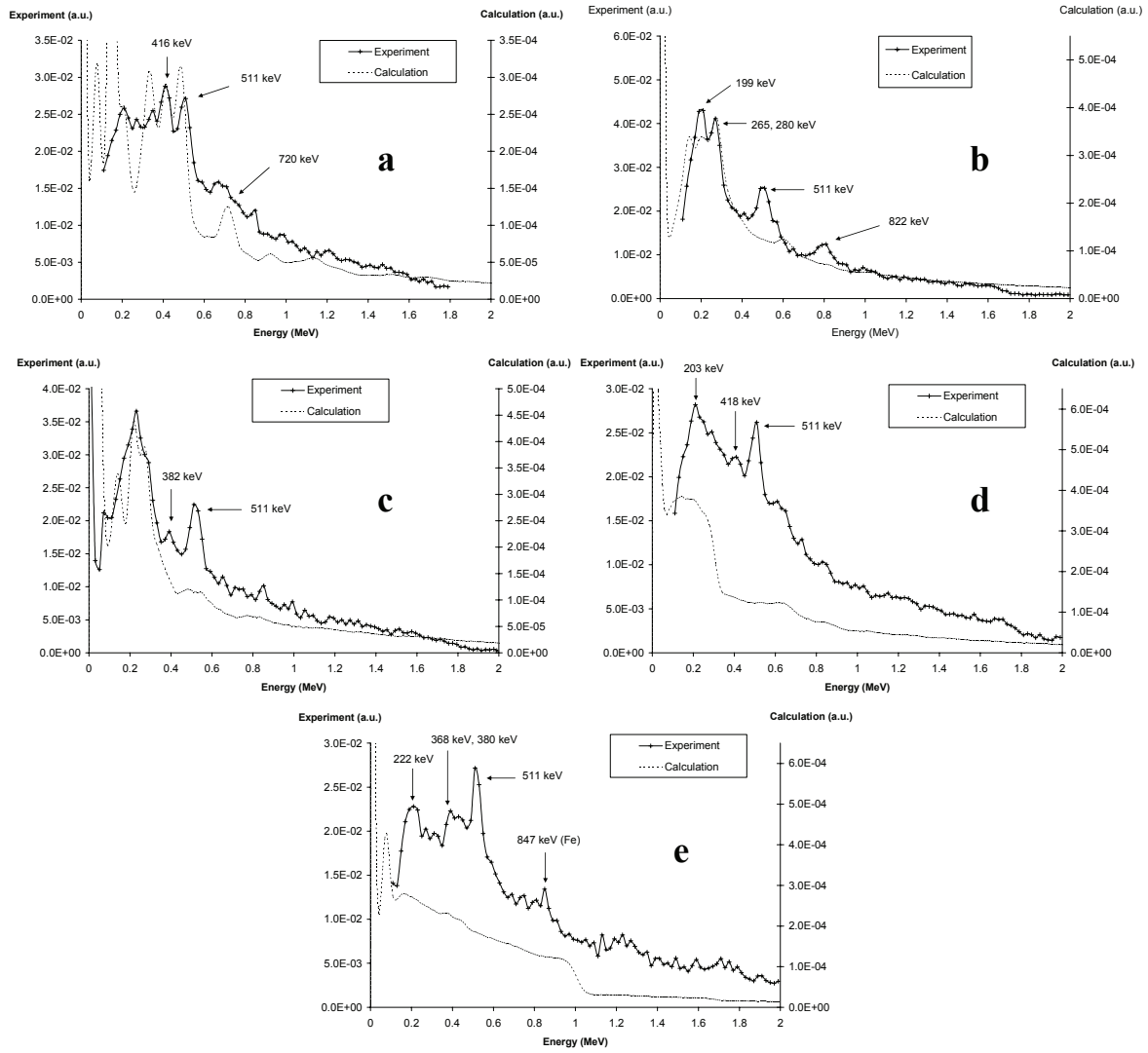

FIG. 1. APT gamma-ray spectra of thallium (a), arsenic oxide (b), bromine (c), iodine (d) and mercury (e).

For thallium, peaks appear at low energy, especially the one near 400 keV that could be associated to a ^{205}Tl transition (2^{nd} to 1^{st} excited levels, 416 keV). Using a better energy resolution scintillator, such as $LaBr_3$:Ce instead of NaI(Tl), would allow to better discriminate gamma rays and thus improve selectivity. However, the cost per volume unit of $LaBr_3$:Ce is still extremely high compared to NaI(Tl), and its larger density (5.08 vs. 3.67 g.cm^{-3}) only partially compensates this drawback, as far as detection efficiency is concerned. For now, to reach a sufficient efficiency at reasonable price on a 0-10 MeV energy range for this application, NaI(Tl) has been preferred [4]. On the other hand, complementary acquisitions will be performed with a high purity germanium (HP Ge) detector showing a much better energy resolution than the NaI(Tl) crystal, to better identify the "peaks" of the APT spectra. The high resolution spectra will be acquired in the frame of the TNA signature measurements (see Section 3), by recording fast neutron induced gamma rays during the pulses of the neutron generator, in complement to TNA gamma rays that are acquired between the pulses.

It can be noticed that HP Ge was discarded for the APT application, mainly because of insufficient detection efficiency and time resolution.
Gamma rays corresponding to transitions between several levels of ^{75}As excited by inelastic scattering reactions, and its fundamental level, can be observed in the arsenic oxide spectrum at 199, 265, 280 (these two last merged in a broad peak) and 822 keV. They could be used to identify arsenic but a definitive conclusion about a practical detection capability would require further tests with a portable setup and small quantities of arsenic, such as those reported in [4] for fluorine, sodium, phosphorus, sulphur and chlorine.
The bromine spectrum shows a small peak at 382 keV associated to the 5th exited level of ^{79}Br. A broad structure centred around 240 keV includes several gamma rays due to the relaxation of excited levels of ^{79}Br and ^{81}Br following inelastic scattering, or excited reaction products such as ^{81}Br(n,2n)^{80}Br. Here also, the use of a $LaBr_3$:Ce detector would improve data analysis. Preliminary acquisitions performed so far with the HP Ge detector (see *FIG.3* in Section 3) confirm the presence of 217, 245, 261 and 276 keV gamma ray transitions.
In the iodine spectrum, small peaks can be guessed at 418, 593, 745 and 828 keV, corresponding to transitions between ^{127}I levels, but no significant gamma ray can be noticed for the detection of small quantities of iodine. It must be reminded that this acquisition corresponds to 3 kg of pure iodine crystals with several NaI(Tl) detectors and an acquisition time of several hours.
The mercury spectrum acquired with a 5kg pure liquid sample also does not evidence significant gamma ray exploitable for the detection of smaller quantities of mercury. A few peaks can be guessed at 222 keV (but very close to the low energy threshold due to self-absorption in the sample), 368 and 380 keV (merged), which could be associated to transitions between nuclear levels of mercury natural isotopes. The peak at 847 keV is due to an iron housing of the mercury bottle.
Monte Carlo calculations performed with MCNPX [8] and a thin target model [5] are also reported in *FIG.1*. It must be mentioned first that this model does not reproduce the 511 keV annihilation peaks, which was expected. For iodine and mercury, however, simulation does not provide a realistic spectrum shape, and even for the other elements, the overall agreement with experiment is not satisfactory. For thallium, models are used in the calculation for because ^{203}Tl and ^{205}Tl data tables are absent of the ENDF/B-VII library. Large peaks are found by calculation near 490 and 720 keV, which could theoretically correspond to transitions between high levels of ^{205}Tl but are not found in experiment. Concerning arsenic, the 822 keV peak is not present in calculation whereas the calculated peak near 600 keV does not appear in experiment. Further tests with the high resolution HP Ge detector will be used to confirm these observations. As to bromine, calculation approximately reproduces the broad structure observed in experiment. However, it must be noticed that ENDF/B-VII lacks gamma-ray production cross sections data and produces no result. Therefore ENDF/B-VI was used, but as previously for thallium, calculation is based on models because no cross section is available for ^{79}Br and ^{81}Br.
The low energy APT spectra of fluorine, sodium, phosphorus, sulphur and chlorine were also recorded, albeit these elements already showed useable peaks at higher energy [4]. Additional gamma rays were observed at low energy, especially a 440 keV peak associated to the 1st excited level of ^{23}Na, and several gamma rays in the fluorine spectrum associated to excited levels of ^{19}F or reaction products like ^{16}N and ^{17}O produced by the (n,t) and (n,α) reactions, respectively. One can particularly mention the 110 keV (1st level of ^{19}F: the electronic threshold was specifically lowered for this acquisition), 197 keV (2nd level of ^{19}F), 298 keV (2nd level of ^{16}N) and 871 keV (1st level of ^{17}O) gamma rays.
As a conclusion, the gamma rays observed in the low energy region will be exploited both to detect elements that do not show peaks at high energy, but also to improve the selectivity and

detection confidence for elements that also have useable peaks at higher energy. Using a low energy threshold in inspection systems dedicated to small objects, where photon attenuation generally remains acceptable down to 200 keV, is extremely valuable.
On the other hand, significant discrepancies were observed between calculated and experimental spectra, as well as non-physical behaviour in a number of calculated spectra. Nuclear databases and consequently numerical simulations are therefore not sufficient to predict the performances of the chemical agent detection system. The acquisition of simple element signatures is compulsory, both for APT and pulsed neutron interrogation.

3. PFTNA signatures

The tests were performed with a PFTNA (Pulsed Fast and Thermal Neutron Analysis) installation available in CEA/DIF. Gamma ray spectra were recorded during and between the pulses delivered by a DT neutron generator, with more than one hour acquisition time and samples of the elements of interest of well-known composition. The decay spectra after irradiation were also recorded during several hours.
The main characteristics of the PFTNA experiment are [7]:

- the pulsed neutron generator producing 14 MeV DT neutrons (GENIE 16T from SODERN) with an average rate of 5.10^7 n/s;
- a Compton Suppression Spectrometer (CSS) composed with a high purity germanium detector with 70 % relative efficiency surrounded by an annular BGO scintillator as veto detector;
- a shield between the neutron source and the CSS to protect the HP Ge and BGO detectors and improve SNR.

The neutron induced spectrum of sulphur acquired during the pulses of the neutron generator is shown in *FIG.2*. The net gamma ray spectrum is obtained after subtraction of the background, which corresponds to the measurement without the sulphur sample. It is worth mentioning that MCNP calculation (not reported here) does not provide the lines associated to (n,p), (n,np) and (n,α) reactions because the corresponding cross sections are not detailed by excited level of the produced nuclei in ENDFBVII and no gamma production table is available.

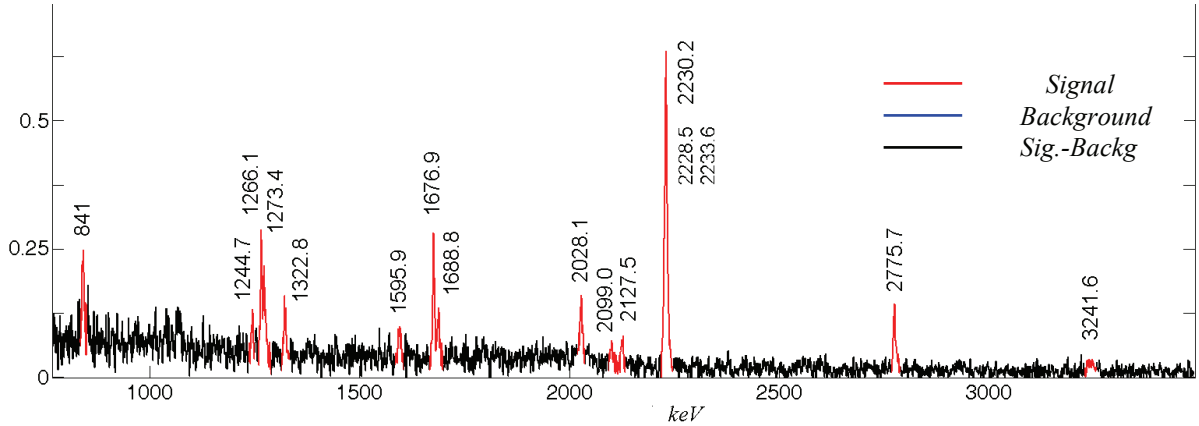

FIG. 2. Gamma-ray spectrum of sulphur measured during the pulses of the neutron generator.

The neutron induced spectra of a potassium bromine sample acquired during and between the pulses of the neutron generator, and after stopping the neutron generator, are shown in *FIG.3*.

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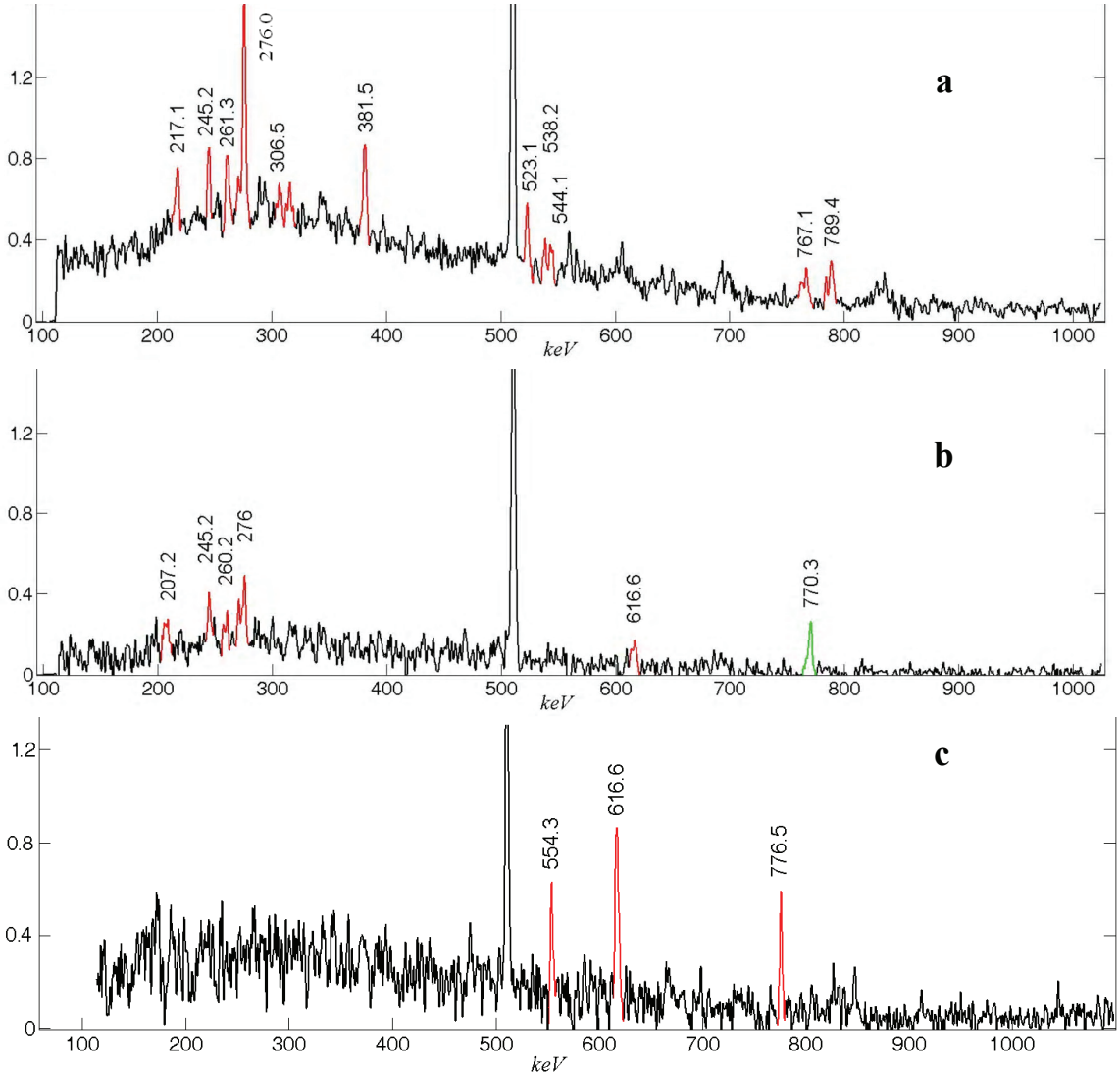

FIG. 3. Gamma-ray spectrum of potassium bromine measured during the pulses of the neutron generator (a), between the pulses (b) and after stopping irradiation (c).

Several fast neutron induced gamma rays are logically observed in the spectrum recorded during the pulses. The 217, 245, 261, 276 and 306 keV lines can probably be associated to the broad structure observed in the bromine APT spectrum of *FIG.1*. The 382 keV peak is also confirmed here. The other lines at higher energy were not clearly observed in the APT spectrum. The gamma rays produced by inelastic scattering (217, 261, 276 and 306 keV) show relative intensities comparable to the 245 keV peak produced by the (n,2n) reaction, which was not expected from the 14 MeV neutron cross sections. In fact, the MCNP calculated neutron spectrum (see *FIG.4*) shows a component near 1 MeV stronger than the 14 MeV peak in our PFTNA device. Concerning the inter-pulse spectrum, the 245 keV peak is due to neutron capture on bromine but the presence of the 260 and 276 keV gamma rays is surprising. The 260 and 276 keV lines can be explained by the long 34.6 µs half-life of ^{81}Br second excited level, from which these gamma transitions are produced. The neutron pulse duration is indeed about 30 µs and the inter-pulse gamma acquisition gate starts immediately after. Concerning the activation spectrum recorded after stopping the neutron emission, clear delayed lines can be observed but the acquisition time was several hours.

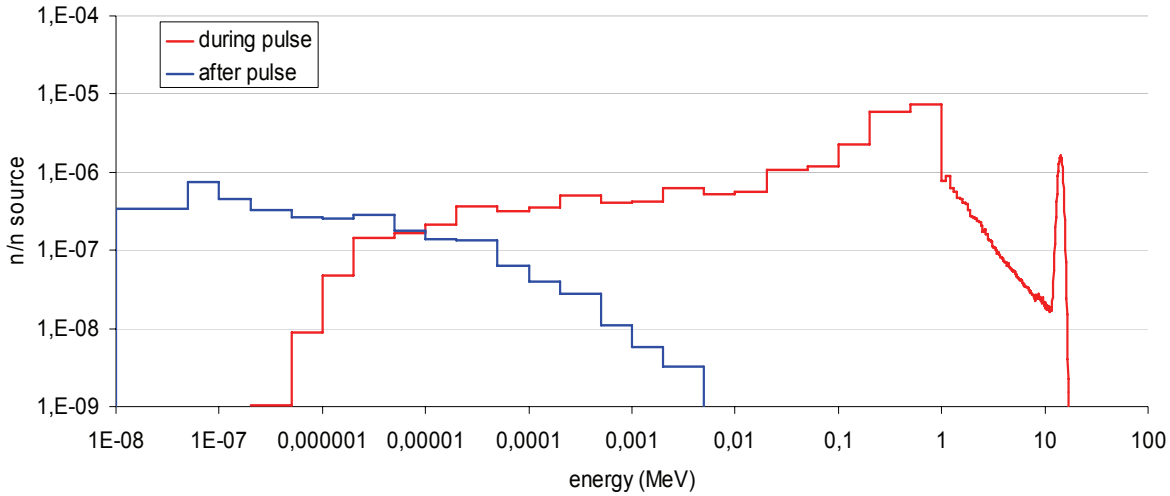

FIG. 4. Neutron spectra calculated with MCNP at the sample position.

7. Conclusion

The gamma-ray spectra acquired for a number of special chemical elements (such as fluorine, sodium, phosphorus, sulphur, chlorine, arsenic, bromine, iodine, mercury and thallium) form an extended experimental database useable for the development of a portable inspection system dedicated to chemical agents in the field of CBRN detection. Gamma rays induced by fast and thermal neutrons were recorded, respectively with the Associated Particle Technique (or APT, in a continuous neutron emission mode) and Thermal Neutron analysis (TNA, with a pulsed neutron generator). In the pulsed mode, fast neutron induced gamma rays were also recorded with a high energy resolution HP Ge detector to help analysing APT signatures recorded with low resolution NaI(Tl) scintillation detectors.

In addition to the specific elements of interest mentioned above, other signatures that must be known to analyse the inspection data are also being recorded: carbon, oxygen, nitrogen, aluminium, iron, chromium, nickel, copper, lead, etc. They will be used to identify other materials potentially found near the chemical agents and to resolve possible peak interferences.

In complement to the above acquisitions performed with large and heavy equipments, tests are underway to demonstrate the detection capability of a portable neutron inspection system for the main elements of interest. The APT potential was already reported for fluorine, sodium, phosphorus, sulphur and chlorine in a previous work [4]. Similar detection tests will now be conducted for thallium, arsenic and bromine, for which this work revealed useable signatures at low energy. Scintillation detectors with a better energy resolution than NaI(Tl), such as $LaBr_3$:Ce, would allow to fully exploit the low energy region that is rich in gamma rays. This crystal is, however, very expensive at present time and this option depends on the potential market of the portable inspection system. Concerning TNA detection tests, a special attention will be paid to iodine and mercury, which are difficult to detect with APT.

Next step is to develop a laboratory mock-up with a light neutron generator, dedicated detectors, electronics and software, so as to test the different options and operating settings for these components, and to establish the ultimate performances of a portable neutron inspection system.

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