DEVELOPMENT OF NEUTRON INTERROGATION TECHNIQUES TO DETECT DANGEROUS SUBSTANCES

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Abstract
The transboundary movement of dangerous substances is being internationally regulated because of the environmental consequences caused by the incorrect management of these chemicals. The international community is committed with a multitude of agreements to control and detect the illicit traffic of such substances in port containers using non-intrusive techniques. Basel and Rotterdam Agreements, Programme 21 are some of the international agreements signed. After initial trials with an accelerator based neutron source, this work reports the following effort to build a reliable experimental data set of the gamma emissions that can, in practise, be effectively recorded from the chemical substances of interest as: mercury, chlorine, cadmium and chromium. When gamma emissions of each chemical element are identified at laboratory scale, the technique will be carried one step forward to test realistic cargo arrangements. Neutron techniques are expected to be used as confirmation tools according as the results of the Coordinated Research Programme (CRP) of the International Atomic Energy Agency (Mayer a et al., 2009).

Keywords: Substances, dangerous, traffic, scanner, neutrons

Introduction
The transboundary movement of dangerous chemicals causes a great worldwide concern. Illegal substances downloaded in not authorized places produce a negative impact on the environment and in human health. As an example, the Minamata Case could be mentioned.
Some relevant substances that are used in the international market are mentioned below.

- Mercury. Liquid metal that rapidly evaporates at room temperature. Methylate mercury presents highly toxicity and characteristics of bioaccumulation and biomagnification in the trophic chain.
- Chlorinated compounds. High toxicity, frequently used as pesticides.
- Chromium. The hexavalent form presents enough scientific studies that confirm its capacity to produce cancer and chromates are toxic to plants.
- Cadmium. Metal that can be accumulated in the trophic chain, cannot be metabolized and is transmitted without changes in the food chain and is a carcinogenic element.

In regard with the internationally signed agreements, it can be mentioned: the Basel Agreement which states that: "the Parties shall cooperate in monitoring the effects of the management of dangerous wastes on human health and the environment". The lines of wastes that shall be monitored are mentioned in its Appendix 1; chlorinated compounds correspond to lines Y10, Y43 and Y44; chromium VI corresponds to line Y21; cadmium corresponds to Y26 and mercury corresponds to Y29; and in Programme 21, chapter 35, mentions the utilization of sciences in support of the prudent use of the environment and the development in support of the daily survival and future development of humanity.

Experimental Method

In order to complement the non-intrusive inspection of goods with X or gamma photons irradiation, neutron techniques could be applied to identify the chemical substances of interest in containers. During the application of this method, different experiments were done in order to identify the prompt and early decay gammas obtained from a (n,γ) reaction induced by an isotopic neutron source and visualize the future application of particle accelerators as a technological feasible neutron source. Different international efforts have been done irradiating with fast neutrons (Buffer, 2004 and Sowerby & Tickner, 2007).

In the first instance, a 25 MeV linear electron accelerator has been employed (Fig. 1) as a pulsed neutron source with a detector array suitable for the detection of the gamma response induced by neutrons incident upon the substances of restricted or dangerous transport. This was initially presented within the frame of Coordinated Research Project of the International Atomic Energy Agency (Position sensitive detection of concealed substances employing pulsed slow neutrons. Second Research Coordination Meeting on Neutron Based Techniques for the Detection of Illicit Materials and Explosives, UN Internat. Atomic Energy Agency Coordinated Research Project) (Mayer b et al., 2007).

Through this means, contents of chlorine were identified and, with an array of fast neutron detectors, samples in the order of tens of grams of uranium-235 were detected (Tartaglione, Di Lorenzo & Mayer, 2009). At a later stage, isotopic neutron sources (^{252}Cf and Am-Be) were employed in order to review the observable gamma responses from the substances of interest. Although the available ^{252}Cf (10^4 n/sec approximately) neutron source is accompanied by a much cleaner gamma environment, its low emission rate resulted in the adoption of the higher intensity 1Ci alpha Am-Be source.

Several tests were carried out in order to optimize shielding and maximize signal to noise ratio (Fig. 2). Under these conditions, gammas induced by the neutron source in the surrounding environment partially obscure the signal from the sample.

As a result of this experimental review implemented with a semiconductor detector, regions of interest for each set of recorded gamma spectra were established as an identification of each tested sample. This has been a necessary step prior to the implementation in a pilot scale scanner with batteries of lower costs gamma detectors.

The irradiated samples were:

- Liquid Hg: prismatic sample 50 mm thick in a glass container.
- Analytical HgO: three 100 g commercial cylindrical containers.
- Analytical Cr: prismatic 400 g sample, 20 mm thick in a glass container.
- Cd: 0,9 mm thick sheet, 35 x 11 cm^2 area.
- Cl: prismatic 1000 g sample, 40 mm thick of sodium chloride.

For the purpose of these measurements, the original fast neutrons were moderated (slowed down) in paraffin and source gammas were shielded with lead. The semiconductor detector employed was a Ge(Li) with moderately high energy resolution.

Although irradiations with fast and epithermal neutrons were also made, in this work only the main results obtained with thermal incidents neutrons are presented. The object of irradiation with neutrons of other energy ranges is to distinguish other useful signals that may help characterize each element.

Results

The identified gamma signals were checked with libraries (Reus & Westmeier, 1983) of typical emissions for each isotope.

In the graphs that follow gamma spectra are shown under two conditions, with the sample present in front of the neutron beam and without the sample. This second case constitutes the background signal (Bg) to be compared with the sample signal.

For the liquid mercury case (Fig. 3), two relevant low energy signals are visible, while the mercury oxide sample (Fig. 4) evinced only the 300 keV signal. This yields confidence to the fact that this last signal arises from the sample and not from any other spurious source, thus coinciding with previous results obtained with the electron linear accelerator as the intense neutron source.

Chlorine is an easily identifiable element due to the important number of different gamma energies it emits when exposed to a thermal neutron beam. Figures 5, 6 and 7 correspond to different energy ranges of a same measurement. There, the energies of the most relevant emissions are shown.

Neutron capture cross sections of chlorine and cadmium are very high for low energy neutrons.

In the particular case of cadmium (Fig. 8) a significant 450 keV emission appears, according to the approximate calibration implemented.

Characteristic signals from chromium have not yet been identified and arsenic has also not given conclusive results to this date.

Fig. 1. Experimental simulated cargo array irradiated with an accelerator neutron source. The neutron beam impinges from the right. The neutron panel detectors appear in each side of the image and on the left, a natural uranium fuel element is visible.

Fig. 2. Experimental set up for semiconductor gamma detector and isotopic neutron source. On the right, the Am-Be source contained in its paraffin moderator. On the left, the Ge(Li) detector surrounded by lead shielding.
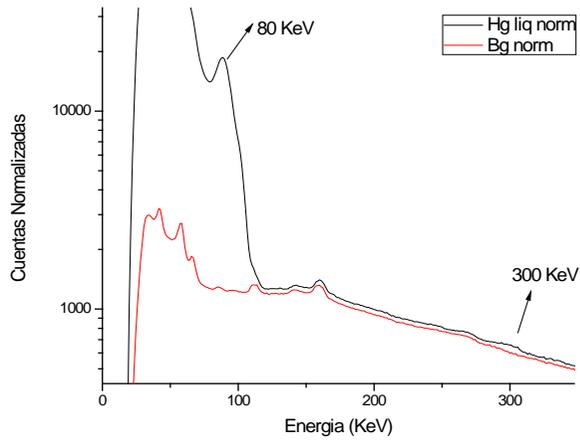
Fig. 3. Gamma spectrum. Sample: liquid mercury.
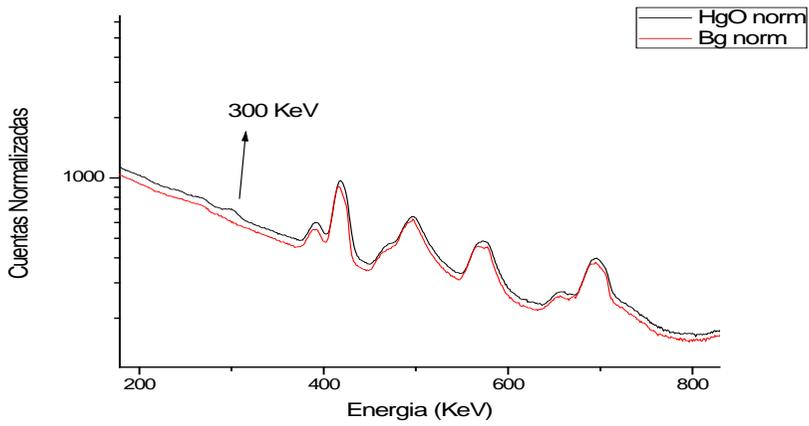
Fig. 4. Gamma spectrum. Sample: mercury oxide.

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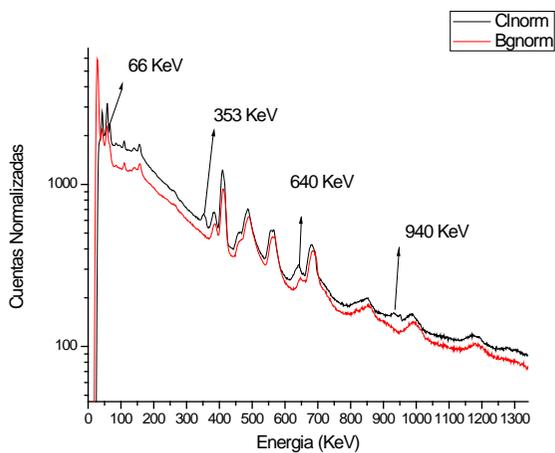
Fig. 5. Gamma spectrum. Sample: chlorine. Low energy.
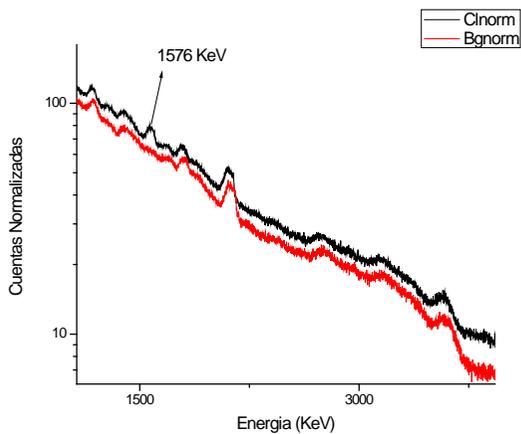
Fig. 6. Gamma spectrum. Sample: chlorine. Intermediate energy.
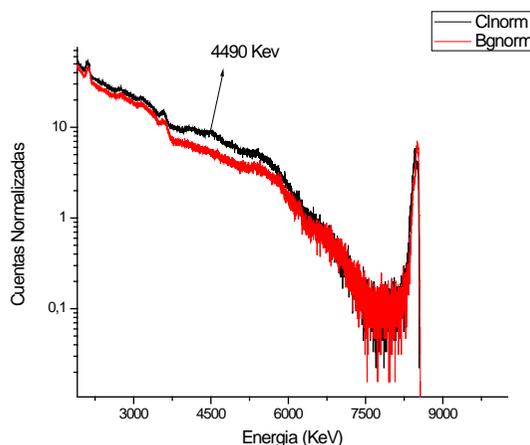
Fig. 7. Gamma spectrum. Sample: chlorine. High energy.
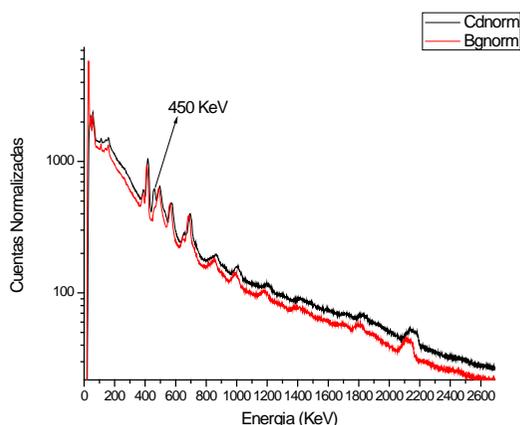
Fig. 8. Gamma spectrum. Sample: cadmium.

Final Remarks

The detection system employed in the preliminary measurements does possess a moderately high energy resolution, but its cost and operative conditions preclude its application in a real scale scanner. In the latter case, a battery of gamma detectors covering a wide solid angle would be mandatory and, thus, lower cost instruments operated at room temperature would become the choice.

Chlorine showed to be an easily identifiable element while mercury and cadmium could be detected with more difficulty in real cargo.

For the purpose of perfecting detection through neutronic tools, measurements with incident neutrons of different energy ranges continue to be done and thus, it is expected to produce a more complete experimental database to help identify substances of interest in the midst of a complex cargo array.

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