NEUTRON RADIOGRAPHY OF CARGO CONTAINERS USING 2.5 MEV TAGGED NEUTRONS

Contact person: Giancarlo Nebbia
Other contributors: Marcello Lunardon, Silvia Pesente, Giuseppe Viesti
Institution: Istituto Nazionale di Fisica Nucleare, Via Marzolo 8, 35131 Padova, Italy
Fax: +39-0498762641 e-mail: nebbia@pd.infn.it

Introduction:
The threat of terrorist use of explosive devices, chemical, biological and radioactive agents has become realistic since the SARIN attack in the Tokyo subway system on March 20, 1995 and after the tragic events of September 11, 2001. The possibility of further attacks against civil populations is one of the most important issues on the international political agenda.

A scenario often evoked implies the use of the so called “dirty bombs”: a sizeable quantity of radioactive material detonated by conventional explosive and dispersed in the environment. Illicit trafficking of explosives and fissile material through the conventional commercial networks (air, maritime and terrestrial) therefore represents a real challenge to security for the future.

Manual and visual inspection of large commercial payloads at terrestrial borders (trucks), airports and seaports (containers) would not be a viable solution both from efficiency considerations and for legal reasons. The standoff inspection of cargo by means of imaging and analytical methods in an integrated system based on a sound technology to identify threat materials is then needed. The key to distinguishing explosives from benign materials is the use of elemental analysis. While x-ray or \( \gamma \)-ray based systems (in particular, CT) can give good precision density measurements with high-resolution three-dimensional images, these systems provide at best only gross information about the elemental content of the inspected item (low Z vs. high Z). Neutron interrogation, however, offers the possibility of measuring the elemental density of most elements in materials.

The use of neutron induced reactions for non-destructive bulk elemental analysis is well documented. All neutrons, in particular fast neutrons, are well suited to explore large volume samples because of their high penetration in bulk material.
Fast neutrons can be produced efficiently and economically by natural radioactive sources, small accelerators or compact electronic neutron generators, making possible the use of neutron based techniques in field applications.

Gamma-rays produced by irradiating the sample with neutrons gives the elemental composition of the material, moreover, knowing the nuclear cross-sections and estimating the absorption factors in the different materials, it is possible to perform a quantitative analysis of elements in the sample even in depth; this is the most suitable technique for detecting hidden explosives. Furthermore with the use of "tagged" neutrons it is possible to determine the local distribution of elements inside the sample volume, or to inspect a precise element of volume (voxel) that has been identified as suspect. Secondary neutrons produced by the irradiation of the sample can be used, by means of multiplicity measurements or spectral analysis, to identify the presence of fissile materials in the inspected volume.

The use of fast neutrons for the inspection of large containers as well as for the screening of check-in luggage in airports has been proposed in the past and there exist a few devices that are now commercially available, although none have yet been deployed for regular operation.

The “PELAN” system developed by Western Kentucky University and now produced and commercialized by SAIC Inc. (USA) is a compact device based on a sealed neutron generator producing about $10^8$ n/sec at 14 MeV and a single $\gamma$-ray counter detecting the gamma quanta produced in the irradiation by neutrons of the investigated object. Such device has the advantage of the compactness and portability (weight is about 45 kg) but it has a severe limitation in applications like container inspection due to the lack of directionality of the neutron radiation produced. As a consequence the gamma radiation collected in the counter originates from the inspected object as well as all the surrounding material evenly irradiated by the neutron flow.

The system produced by RAPISCAN (USA) is a much larger device specifically designed for cargo inspection. It has a good level of directionality since the neutron flow is collimated into a fan beam, and it makes use of a large number of $\gamma$-ray counters located around the inspected volume which are collimated as well. The severe drawbacks of such system consist in the complexity and cost of the device and in the use of a large and expensive particle accelerator needed to provide nanosecond time structure to the neutron fan beam.

A more compact system based on the so called “associated particle technique” has been successfully built and tested in the framework of the EURITRACK project financed by the European Union under the 6th Framework Programme. This method allows to tag neutrons in
time and direction instead of trying to create collimated neutron beams from sources that are, by their nature, isotropic.

It goes without saying that, when analyzing spectroscopic data originated from interactions of tagged neutrons with a sample, the gain in signal/noise ratio is remarkable, improving sometimes by two orders of magnitude.

On the basis of past experience on the production of tagged neutron beams of 14 MeV troughs the “associated particle technique” we proposed then to extend the study of such technique with 2.5 MeV neutrons produced in the d + d reaction. The energy of about 800 keV of the $^3$He recoiling nucleus associated with the neutron makes the detection more critical. On the other hand the lower energy of the neutron beam would supposedly induce a much lower background rate when traversing a sample, in particular a fully loaded container.

We proposed a full investigation of the technique including the design and construction of $^3$He and neutron detectors, the collaboration with neutron generator manufacturers and the installation of laboratory test equipment to produce results on neutron attenuation measurements across a sea container under different load conditions.

Due to financial difficulties arisen a few months after the beginning of this CRP, our group switched from the construction of a system based on a new kind of neutron generator to a simpler one based on the use of a tagged radioactive neutron/gamma source. The main trends of the project’s objective remained unchanged, and the results are rather satisfactory.

The original objectives included:

a) To provide a method to inspect containers by “low resolution neutron attenuation imaging” making use of the low background conditions provided by neutron tagging.

b) To study the coupling with a standard gamma attenuation technique to provide “low resolution elemental imaging” of the container content

Both of these objectives have been essentially preserved and reached with the new setup we have implemented. One has to underline the fact that, as it will be discussed in detail below, the loss of directional tagging of neutrons (preserving only the time tagging) does not seem to reduce significantly the overall improvement on the signal/noise ratio.

Material recognition is based on the atomic number Z dependence of the relevant photon absorption coefficients: it is a well established method at low photon energy, where the photoelectric effect dominates, while it becomes critical for increasing photon energy as is
required in order to increase the penetration of radiation to inspect thick objects. In fact, when the photon penetration is not sufficient the object’s image appears black thus becoming one of the important issues in security inspection.

In all cases when photon irradiation is unable to disentangle the problem of inspections, the use of neutrons as probing radiation has been often proposed. To this end sophisticated techniques have been developed in order to enhance material recognition, especially for low-atomic-number materials, in an effort to optimize the detection of explosives and drugs in customs operation. Examples of such developments are represented by the “combined fast-neutron and gamma-radiography” and the “fast neutron resonance radiography” or by detection of neutron induced gamma rays. In the latter case, gamma rays are utilized to characterize carbon, oxygen and nitrogen nuclei, which are the major components of explosives or narcotics. Discrimination between threat materials and common goods is generally obtained by measuring relevant elemental ratios as C/O and C/N. Some systems, based on the neutron-in/gamma-out technique, are already operational or commercially available. Because of the gamma ray production cross sections (of the order of few hundreds millibarn), such systems require at the moment a rather long inspection time (5-10 minutes) and are thus adequate only to detect rather large quantities of threat materials (several kg). On the other hand fast neutron total cross sections are typically in the region of the few barns, therefore transmission measurements, exploiting all the possible neutron interactions, are certainly more appealing. A device based on this technique has been designed by CSIRO and it is now in operation at Brisbane airport in Australia. The system performs material recognition by transmission measurements of 14 MeV neutrons (produced by a D+T generator) and gamma rays from an intense $^{60}$Co radioactive source. The transmitted radiation is recorded in a detector array made of several individual pixels, providing a rough “elemental picture”. The scanner thus produces a 2-D radiographic image in false colours in which each pixel brings material information. Limitations of this technique are mainly due to its application to rather large aircraft containers in which averaging over all the materials crossed by the radiation represents a severe smearing of the information. Moreover there is no specific signature delivered that identifies directly the presence of C, N and O nuclei in the material.

In order to improve material recognition identifying single elements in the sample, it has been proposed to measure neutron transmission at different neutron energies corresponding to the resonance region in the total cross section for a number of key light elements. Such technique should provide element identification as with fast neutron induced gamma ray
spectra. However, it involves the use of a number of mono-energetic neutron beams or an intense white neutron source with non-negligible technological and licensing problems.

The experimental set-up employed in this project consists of a $10^6$ fissions/s $^{252}$Cf source tagged by requiring the coincidence between two 2” thick 4” diameter NE213 liquid scintillator cells mounted face to face around the source. The burst of neutrons and gammas emitted during each fission event has been thus used as start signal. The efficiency of the tagging system is not relevant to the results shown in this paper; however it is worth mentioning that it was close to 100%. The tagged radiation was first collimated by a 44 cm long lead-polyethylene collimator and then hit the sample placed at the collimator exit. Finally, a third 2” x 4” NE213 liquid scintillator cell was placed at a distance of about 160 cm from the tagged $^{252}$Cf source to detect transmitted neutrons and gamma rays. The low energy threshold of the neutron detector was measured to be about 100 keVee.

The resolution of the time-of-flight system was about $\delta t=1.5$ ns [FWHM] for gamma-gamma coincidences determined using a $^{60}$Co source. For the $^{252}$Cf source the time resolution of the system was slightly worse, about $\delta t=2.0$ ns [FWHM]; this is due to events where the tagging signals were given by neutrons (that have a large natural jitter) introducing a spread in the time-of-flight start.

It is well known that low Z elements as C,N,O, that are the basic constituents of organic compounds, exhibit strong resonances in the neutron cross section. Such resonances generate absorption dips at precise energies in the neutron spectrum emerging from the samples, when the transmission is measured by using a white neutron spectrum. Consequently, the analysis of the transmission as a function of neutron energy can be used to identify the nuclei in the sample. Alternatively, the transmission can be measured using mono-energetic neutron beams with well defined kinetic energies corresponding to the major resonances in the elements to be identified. Despite the large effort done in this field in the last decade, it is worth mentioning that the application of the neutron resonance radiography to the inspection of air cargo was strongly criticized in the past.

Californium sources offer the possibility to look for fast neutron resonances in the range $E_n=0.2$-5 MeV, limited only by the lower energy threshold of the neutron detector and the relatively low yield of high energy neutrons. Moreover, the time-of-flight resolution of the detection system sets additional constraints on the detection of narrow resonances. Nevertheless C,N,O nuclei show a number of sufficiently wide resonance or bump-like structures that can be used to derive signatures of a particular nucleus.
The experimental transmission is obtained as a function of neutron energy as derived from the measured time of flight for graphite, nitrogen and oxygen (water) samples. In each case, the total neutron cross section is reported, taken from the ENDF-BVII library. The nitrogen spectrum was obtained by irradiating a bottle filled with LN$_2$ from which the empty bottle spectrum was subtracted. The oxygen spectrum was obtained measuring a water sample. It has been thus possible to determine, for each element, the energy regions where the relevant resonant structure shows up. This energy region is between $E_n=2.5$-$4.5$ MeV for carbon, which includes the resonances at $E_n=2.814$, 3.52 and 4.27 MeV. In the case of oxygen (water sample), a first region is located between $E_n=1$ and 2 MeV, which include a number of unresolved resonances; the large bump in the cross section at $E_n=3.5$ MeV is also seen. In the nitrogen case, there is again the region of the resonances in the range $E_n=1$-$2$ MeV and then a large structure at $E_n=4$ MeV.

It is interesting to notice that, despite the overlap of the resonance region between the nuclei, the gross shape of the transmission versus neutron energy identifies in a clear way a given nucleus. Information about the density of nuclei inside a given complex sample can be thus obtained by fitting the experimental spectrum with a library of elemental spectra.

We applied the above considerations to a paper sample where we tried to deduce the relative composition of light elements from the transmission spectrum of the sample. The spectrum has been fitted using a weighted sum of the elemental C (graphite) and O (H$_2$O) spectra searching the weight parameters that minimize $\chi^2$. The result of this best-fit yields a C/H$_2$O ratio of about 1, corrected for density effects, in agreement with the expected value. In fact the chemical formula of cellulose (C$_6$H$_{12}$O$_6$) used for commercial paper indicates a 1 to 1 ratio between the carbon and water contribution to the transmitted neutron spectrum. Consequently, we can take this as a demonstration of the possibility to use the time-of-flight distribution to derive the chemical composition of an organic sample.

As for the use of combined information of neutron and $\gamma$-rays absorption, a gamma ray detector was placed at a distance of about 160 cm from the tagged $^{252}$Cf source. The detector used was a 3”x 3” NaI(Tl) scintillator crystal equipped with a Compton-suppression shield to improve the full-energy response. The peak-to-total ratio was indeed measured to be about 70% for a $^{60}$Co source (to be compared with the standard 30-40% value). The time of flight measured between the NaI(Tl) scintillator and the two tagging detectors was used to select gamma rays, thus rejecting events induced by fission neutrons.

A large number of samples have been employed in the data taking, including mono-element samples from graphite to bismuth as well as a set of commercially available substances.
For a given sample of the element $Z$ with known thickness $t$, we measured the transmitted spectrum $I_{\gamma}^{\text{exp}}(E_{\text{det}},Z)$. From the un-attenuated spectrum $I_{\gamma,0}(E_{\text{det}})$, measured with the same detector, and using the NIRST database of attenuation coefficients, it was possible to predict the transmitted spectrum for a sample of the element $Z$ and thickness $t$ by using the well known formula:

$$I_{\gamma}^{\text{calc}}(E_{\text{det}},Z) = I_{\gamma,0}(E_{\text{det}}) \exp [-\mu(Z,E)t]$$

where the attenuation coefficients at $E = E_{\text{det}}$ were used without taking into account possible bias due to the detector response function. The predicted transmission spectrum for the corresponding element fitted nicely the experimental one, with the exception of the range $E<0.5$ MeV, where a low energy peak appears for high $Z$ samples.

Assuming that the element inside the sample is not known, one can compare the measured transmitted spectrum $I_{\gamma}^{\text{exp}}(E_{\text{det}},Z)$ with a library of predicted spectra, $I_{\gamma}^{\text{calc}}(E_{\text{det}},Z)$, and then calculate the reduced chi-squared $\chi^2 / \nu(Z)$ over a defined energy range $(E_1, E_2)$ providing the recognition of the sample material. It is worth noting that the difference in the attenuation of photons of a given energy $E$ for a sample of thickness $t$ of two neighboring elements $Z$ and $Z+1$ depends not only on the change in the mass attenuation coefficient $\mu/\rho$ but also on the change on the density of the materials. Consequently, the sensitivity of the method is much larger for the elements for which the density is strongly varying with $Z$.

It is interesting to note that the possibility of material recognition using photons is strongly correlated not only to the role of photoelectric effects that depend on $Z^5$, but also to the variation of the material density in the vicinity of the element to be identified. Selecting the photon energy range is therefore one of the crucial parameters in order to enhance material recognition, being the low energy region the most sensitive due to the dominance of the photoelectric effect.

A second interesting point that emerged from this project, due to the availability of photons with a wide energy distribution, is the dependence of the material recognition on the photon energy. This is a crucial point in few key applications as the security ones, when the photon energy is increased to obtain high quality images of thicker object. The increase of the photon energy decreases the relative role of the photoelectric effect making the task of material recognition more difficult.

This effect has been studied directly with our experimental data on Ta. In particular, the experimental data relative to the 10.5 mm Ta foil have been analyzed by constructing $\chi^2 / \nu(Z)$ versus the assumed atomic number $Z$ for different energy ranges: 0.5-1.5 MeV, 1.5-2.5 MeV,
2.5-3.5 MeV, 3.5-4.5 MeV and 4.5-5.5 MeV. For each of them, the minimum has been analyzed to extract $<Z>$ and $\delta Z$ values. Results clearly demonstrate that the resolving power of the system worsens when the photon energy is increased.

As for the application of this technique, it is interesting to consider that with the simple setup described one can use the variable energy gamma absorption associated with the neutron/gamma energy integrated absorption ratio and with the resonance neutron absorption to measure different quantities on the same sample and fuse them together to obtain a better material analysis.

It is worth mentioning that the technique proposed here is working very well for a large set of elements, and it seems that it can be applied also in a number of industrial applications for the control of composite materials.