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Detection of special nuclear material with a transportable active interrogation system*

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Abstract. Special nuclear materials hidden in shipping containers are extremely difficult to detect through their faint spontaneous emission of neutrons and photons. R&D efforts focus on active interrogation (AI) techniques, employing external beams of neutrons or high-energy X-rays to first trigger fission reactions and then detect prompt or delayed neutrons and/or photons. Our group created a complete active interrogation system based on detectors developed by the universities of Pisa and Yale and on an ultra-compact linear accelerator (LINAC). The detectors contain liquid droplets that vaporize when exposed to fast neutrons but are insensitive to X-rays. The X-ray generator is based on 9 MeV electron LINAC developed by S.I.T. Sordina S.p.A. for intraoperative radiotherapy. The latter is a standing-wave design that does not require external solenoids for electron radial focusing. Copper is used both as X-ray production target and as collimator, which prevents the production of photo-neutrons. In our first tests, we detected depleted uranium, while excluding significant production of contaminant photo-neutrons.

1 Introduction

A key aspect in the effort to ensure national security is interdicting special nuclear materials (SNM), *i.e.* U-235, Np-237 and Pu-239, from being introduced into our countries, hidden in the large containers entering through shipping ports and carrying the large majority of cargo. The possible presence in these containers of weapon components comprising special nuclear materials is extremely difficult to detect through their faint radioactive signature. While radiation emitted by Pu-239 may be recorded by high-sensitivity devices, that of highly enriched U-235 (HEU) is virtually impossible to detect with passive interrogation techniques. In fact, the emission consists of an extremely low yield of neutrons and a weak emission of low-energy gamma rays which are strongly attenuated by surrounding materials. HEU is not only harder to detect, but possibly also easier to obtain than Pu-239, and thus lends itself to improvised nuclear devices. For these reasons, active interrogation techniques (fig. 1), using beams of neutrons or high-energy X-rays to trigger fission reactions are considered the only viable option to detect the presence of HEU. These techniques are also effective in the detection of Pu-239, as illustrated in a comprehensive DOE report [1] and in the reviews by Slaughter *et al.* [2] and by Medalia [3].

Historically, since the late 1960s [4], active interrogation for the detection of special nuclear materials has relied on the detection of delayed fission neutrons by means of moderator-type neutron detectors. These detectors can be blinded by the intense pulse of prompt gamma radiation that accompanies the fission process. In recent versions of the technique [5], a pulsed beam is used and the detectors are gated off for a few milliseconds during and after the interrogation pulse, in order to allow them to recover from saturation. After this time, the moderator-type detectors can be used effectively to record delayed neutrons emitted with a half-life of up to 1 min after the fission, (see, *e.g.*, [6]).

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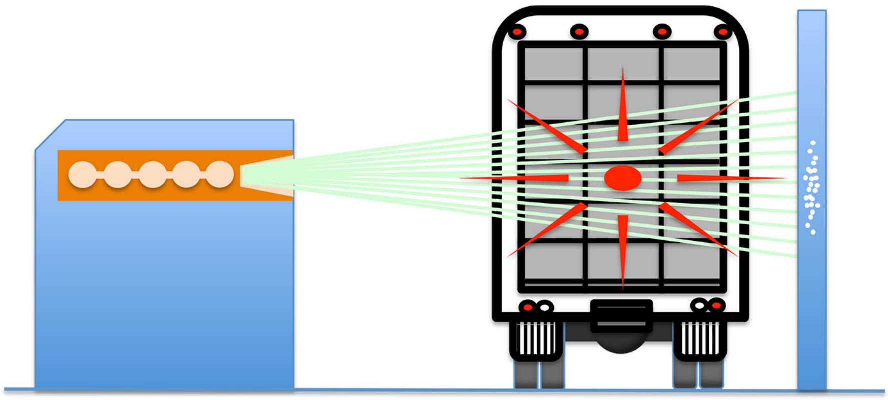

Fig. 1. Schematic of active interrogation system: an external beam of X-rays or neutrons (left-hand side) triggers the emission of fission neutron from special nuclear materials detected by a large detector (right-hand side).

These neutrons carry a signature that is characteristic of special nuclear materials; however, both their emission energies and their yields are relatively low, which leads to a limited neutron flux emerging from cargo containers.

Alternative approaches have been developed relying on the detection of delayed high-energy gamma rays. These gamma rays are produced with yields about 10 times larger than those of delayed neutrons, are not attenuated as intensely, and also carry a signature characteristic of special nuclear materials [7]. While providing a higher sensitivity, the technique still requires intermittent operation of the interrogation system, alternating between irradiation and detection. For improved duty cycle, novel active interrogation techniques focus on the detection of prompt fission neutrons, whose yield is over 100 times higher than that of delayed neutrons. A relevant implementation uses xylene-based liquid scintillators in conjunction with a low-energy neutron interrogation beam [8]. The system is highly sensitive to photons and requires advanced laboratory electronics for pulse-shape analysis and for the identification of the neutron signal against the prompt fission-gamma flash.

This work explores the potential of a complete AI-system based on an ultra-compact linear accelerator (LINAC) and on detectors developed in collaboration between the universities of Pisa and Yale. The system does not require complex electronics or special training in order to be operated and offers the possibility of simultaneous irradiation and detection, *i.e.* a 100% duty cycle. In fact, it relies on an extremely well-established accelerator technology [9,10] and on detectors with an inherent threshold behavior and photon-insensitivity [11,12] in order to provide a "yes or no" answer as to whether prompt fissions are triggered by the active interrogation of a container.

2 Materials and methods

2.1 Neutron detector

The interdiction of special nuclear materials places heavy performance requirements on the detector systems. As mentioned earlier, radiation beams are used to trigger fission reactions, then prompt and/or delayed fission neutrons and/or γ-rays are detected. Among the favored active interrogation approaches is using X-rays from 9 MV electron linear accelerators.

These "9 MV" X-rays have an effective energy triggering adequate photo-fission in SNM (fig. 2), while avoiding neutron production in most "innocent" materials, such as legitimate contents and structural materials of shipping containers. The photo-neutron production threshold for these materials is typically above 10 MeV. An exception is the production of photo-neutrons in naturally occurring deuterium; these neutrons can reach 3 MeV when produced by 9 MV X-rays. Therefore, in order to record only the intense prompt neutron emission, an ideal detector should not only discriminate X-rays but also neutrons below about 3 MeV. Since the scan must be acquired and evaluated in real time, the detectors should be active and offer a rate-insensitive read out.

Superheated emulsions satisfy these requirements [13]. They are suspensions of superheated halocarbon droplets in an inert medium; following irradiation with fast neutrons, these detectors develop bubbles (fig. 3), which can be recorded with various methods and then quickly reset to the liquid phase by pressurizing the detectors. Neutron-induced charged particles generate vapor cavities inside the droplets; when these cavities reach a critical size, the expansion becomes irreversible and the whole droplet evaporates. The amount of energy and the critical size required for bubble nucleation depend on the composition and on the degree of superheat of an emulsion. By appropriate choice of the detector manufacturing and operating parameters, a selective response can be achieved to different types of ionizing radiations [12].

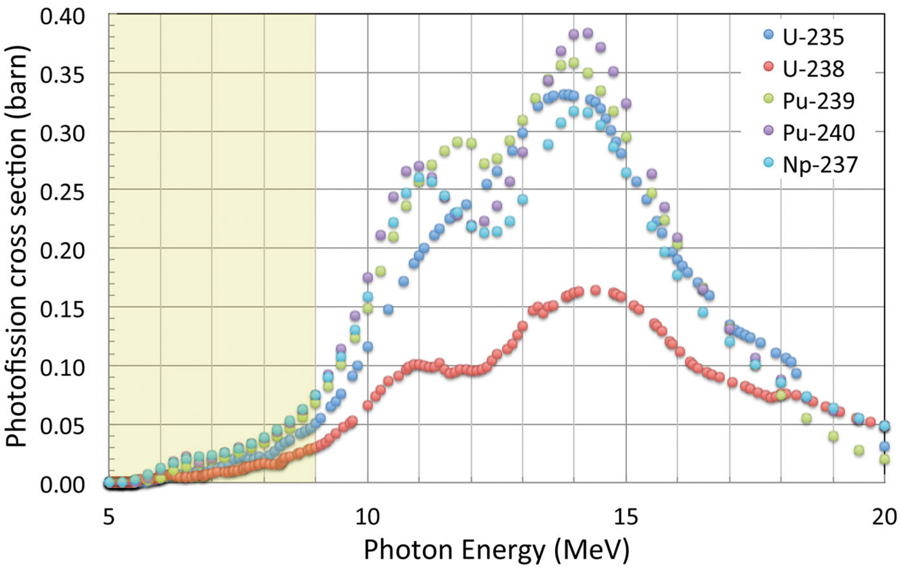

Fig. 2. Photo-fission cross-sections for special nuclear materials of interest. The shaded area indicates the energy range of 9 MV X-rays typically employed for active interrogation.

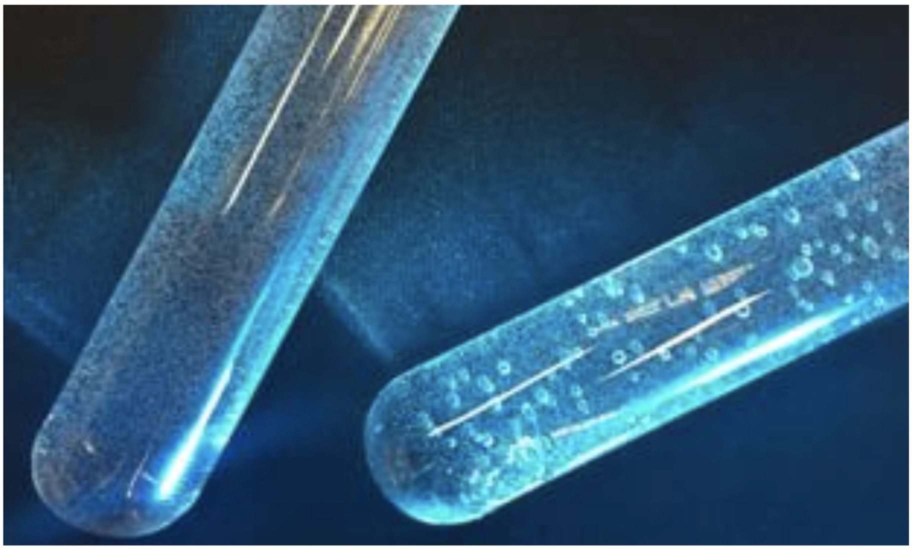

Fig. 3. Superheated emulsions of halocarbon C-318, before (left-hand side) and after (right-hand side) irradiation with fast neutrons.

The detectors are manufactured in the form of emulsions placed inside glass containers, ranging from few-mL cartridges to several-liter tempered glass vessels. The metastable state of a superheated liquid is normally fragile and short-lived due to the microscopic particles and/or gas pockets present at the interface with container surfaces. However, fractionating a liquid into droplets and dispersing them in an immiscible fluid creates perfectly smooth spherical interfaces, free of nucleating impurities or irregularities. Thus an emulsified superheated liquid may be kept in steady-state metastable conditions.

Superheated emulsions have become well-established among neutron detectors and they are included in recent standards issued by ANSI [14] and ISO [15]. Several laboratories manufacture these detectors worldwide and some make them available commercially (`http://bubbletech.ca/`) or within research collaborations (`http://ocr.yale.edu/`). The detectors can be read out either post-exposure, *e.g.*, with commercial image acquisition devices, or in real time, *e.g.*, with dynamic light scattering techniques [16,17].

Number, size and composition of the droplets can be varied in the formulation of the detectors, and this permits a wide range of applications [12]. For example, highly superheated halocarbons can be used for the detection of sparsely ionizing radiations, such as photons and electrons. In this work, halocarbons with a moderate degree of superheat were used (table 1), since they are only nucleated by energetic heavy ions, such as those released by fast neutron interactions

Table 1. Halocarbons used in the superheated emulsions for SNM interdiction.

Halocarbon name and code	Chemical formula	Boiling point (°C)	Critical point (°C)
Octafluorocyclobutane, C-318	$C_4\,F_8$	−7	115.2
Decafluorobutane, R-610	$C_4\,F_{10}$	−1.7	113.3

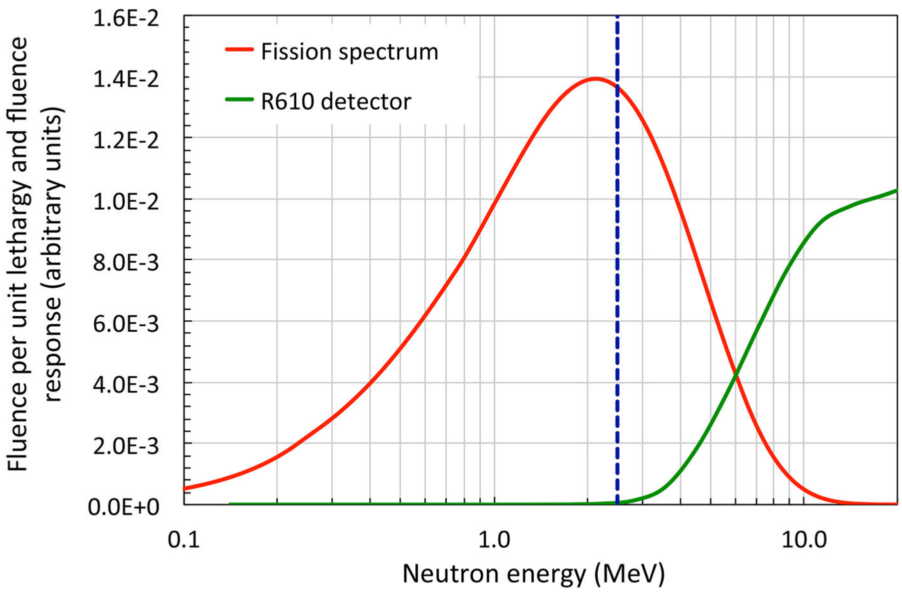

Fig. 4. Fluence response of R610 emulsions *vs.* typical fission spectrum. The vertical dashed line indicates the threshold required to discriminate photo-neutrons from deuterium.

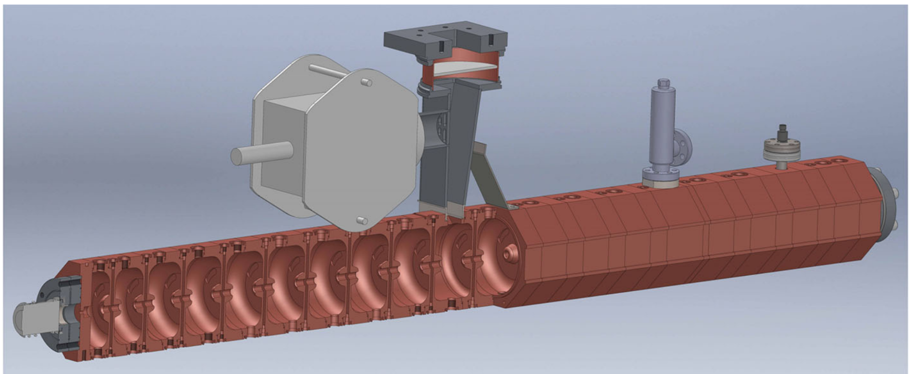

Fig. 5. Schematic 3D view of the SIT-LIAC© linear accelerator tube used in this study.

(fig. 4). In our approach, bubbles are detected and counted with an optical readout approach: when suitably chosen beams of light cross the emulsion, they undergo measurable attenuation and scattering which is detected by arrays of photodiodes placed in contact with the detectors.

2.2 Linear accelerator

A 9 MV light, compact and mobile X-ray generator for active interrogation of cargo containers was designed and assembled in a close collaboration with the company SIT Sordina S.p.A (Vicenza, Italy, `http://www.soiort.com/en/`). The solution for the active irradiation system is based on the SIT-LIAC© machine (510(K) number K110840), which is in use at several hospitals worldwide (fig. 5). The generator uses a linear accelerator (LINAC), whereby an electron beam is produced from a small thermionic cathode and accelerated up to 9 MeV. Radial focusing of the electron beam is achieved electrostatically, therefore no external solenoid is required. This allows for an extremely compact and light LINAC system, with virtually no radiation leakage from the accelerating guide.

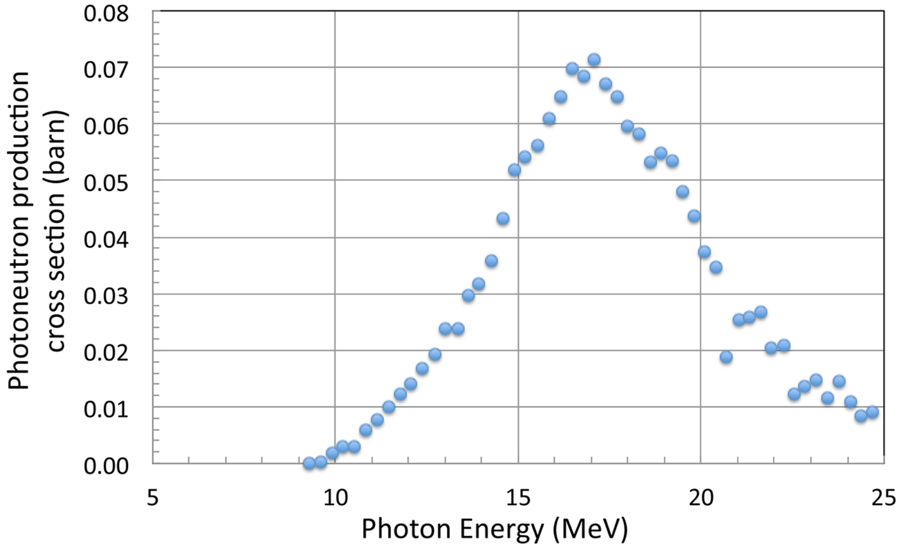

Fig. 6. Cross-section for photo-neutron production in copper showing that X-ray energies higher than 9 MeV are required to trigger the reaction.

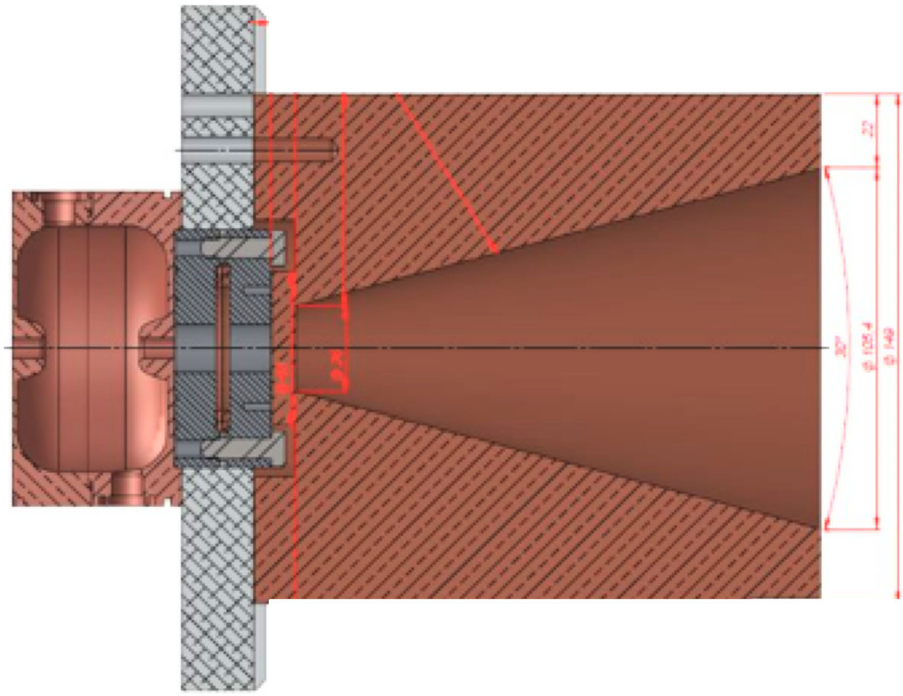

Fig. 7. Technical drawing of the copper assembly for the production and collimation of the beam of X-rays.

A custom-designed X-ray production target was built to generate an intense and clean beam of photons, without neutron contamination. The material of the target was high-purity (beryllium-free) copper, whose photo-neutron production cross-section is nil below 9 MeV, as shown in fig. 6. The X-ray beam collimator was also built with high-purity copper, forming a single piece with the target. This ensured efficient heat removal during the generation of X-rays. Based on prior studies, for our first prototype we chose a target thickness of 4.5 mm, and a conical internal geometry of the collimator with an angle of aperture of 30° (fig. 7).

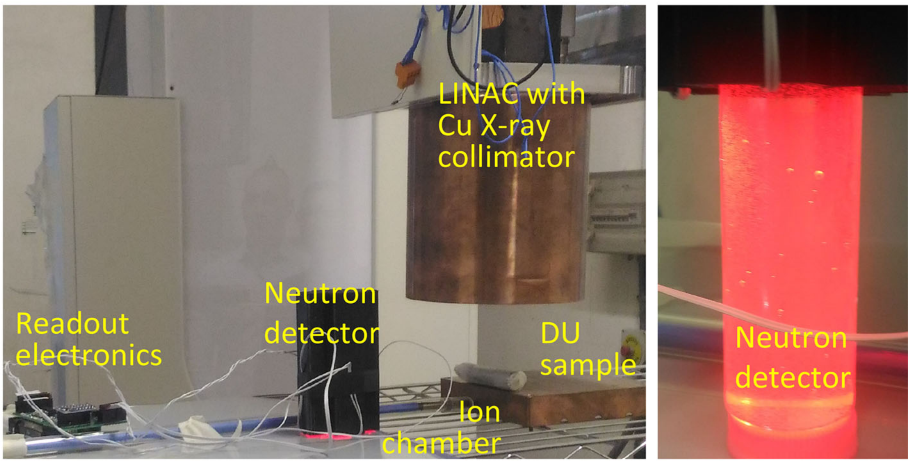

Fig. 8. Experimental set up for the detection of special nuclear material; shown on the right-hand side is the neutron detector with a few bubbles inside, which scatter red light.

Table 2. Stray radiation levels around the LINAC normalized to in-beam ion chamber readings.

Angle [°]	Value [%]
90	< 0.4
135	< 0.3
180	< 0.0003

2.3 System characterization and performance

A series of experiments was carried out in order to characterize and validate our active interrogation system. First, we investigated the radiation leakage levels around the LINAC, as this is a key aspect of its viability [18]. To determine the X-ray leakage radiation, the accelerator tube was placed horizontally at a height of approximately 170 cm from the floor. A Semiflex cylindrical ionization chamber (PTW Freiburg GmbH, Germany) was placed in the beam at a distance of 1 m from the collimator to monitor the output of the accelerator, and two survey meters (models Inovision 451B and 451P; Fluke, Eindhoven, The Netherlands) were placed at several angular positions (90°, 135° and 180°) to map the stray radiation field around the LINAC.

Next, we tested the system in terms of its ability to trigger and detect photo-fission in special nuclear materials, while producing negligible amounts of photo-neutron contamination. Tests were carried out with and without a sample of military-grade depleted uranium inside the X-ray beam (fig. 8). A neutron detector based on emulsions of R610 was placed in the proximity of the X-ray beam. The detector was operated at atmospheric pressure and at a temperature of 19 °C, these operating parameters correspond to a 3 MeV neutron threshold. The X-ray radiation levels next to the neutron detector were monitored with the Semiflex ionization chamber.

3 Results and discussion

The results of the radiation leakage measurements around the linear accelerator are given in table 2 and shown as a polar diagram in fig. 9. The distribution of radiation levels indicates that the only source of stray radiation is the target itself, while the accelerating waveguide presents virtually zero leakage.

While the measured leakage radiation levels are compliant with safety standards for radiotherapy equipment, an even lower radiation leakage may be easily achieved with some additional shielding around the copper collimator. Namely, a 1 cm thick tungsten sleeve would reduce the 0.4% leakage measured at 90° down to 0.04% with an additional weight of only 14.5 kg. This may be of interest in scenarios where the accelerator must be operated by personnel in its close proximity.

The photo-fission tests were equally satisfactory. As shown in fig. 10, when a depleted uranium sample was placed inside the 9 MV beam of X-rays, we could clearly observe the progressive formation of bubbles, through the measurement of scattered light. When the DU sample was instead removed and the beam was turned on, the detector did not record any counts. No response was observed even when some hydrogenous materials were placed in the beam, such as plastic and wood samples containing naturally, occurring deuterium. Photoneutrons were produced by the 9 MV X-ray beam through the photodisintegration of deuterium, but these neutrons were below 3 MeV and they were effectively discriminated by our detector threshold.

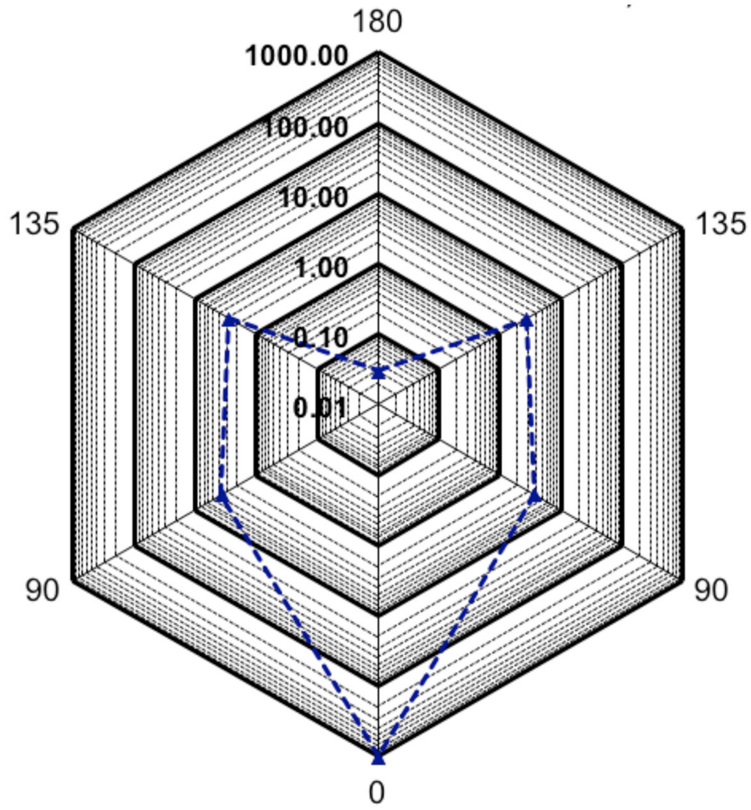

Fig. 9. Polar diagram of the leakage radiation levels around the LINAC used in this study.

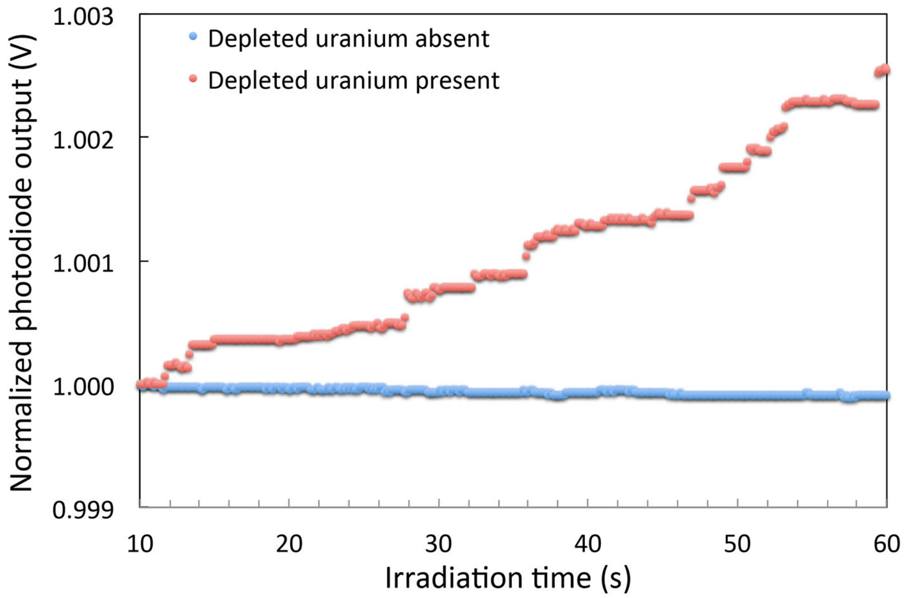

Fig. 10. Response of R610 emulsions when the 9 MV X-ray beam is on and a sample of depleted uranium is either present or absent inside the X-ray beam.

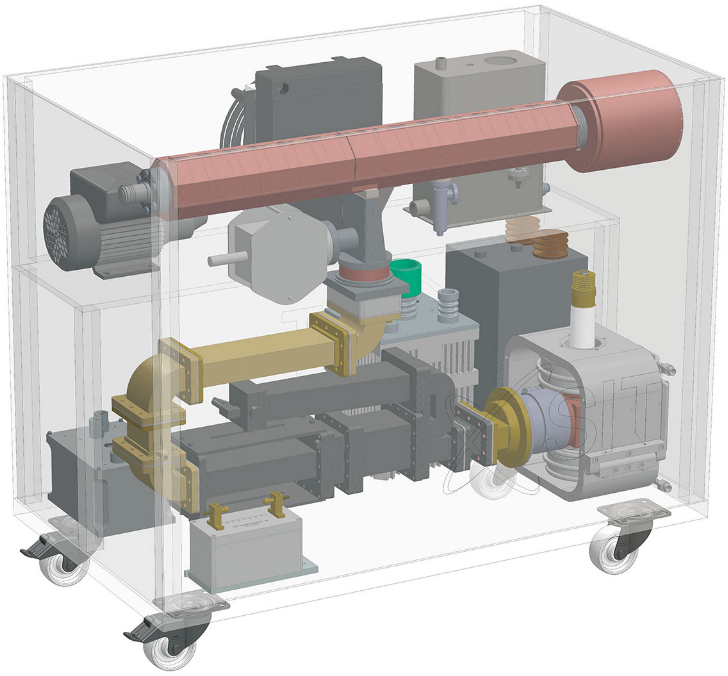

Fig. 11. Preliminary layout of a dedicated LINAC for active interrogation.

In conclusion, our experiments proved that our system can trigger and detect the presence of special nuclear materials. Our X-ray generator does not produce photo-neutrons, while those produced by the photodisintegration of naturally occurring deuterium are effectively discriminated by our detectors. We have designed a preliminary layout of a proposed active interrogation system, as shown in fig. 11. Our design minimizes both power requirements and the mass of shielding needed for a safe operation: LINAC, modulator, cooling unit and RF power supply are all assembled in a single enclosure. These features make our proposed system extremely compact and unique in its class: the expected size is about $0.5\,\mathrm{m}^3$ while the expected weight is $< 200\,\mathrm{kg}$. The entire apparatus is so light and compact that its implementation appears possible in fixed and mobile (terrestrial, maritime and even aerial) interrogation systems.

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