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Contributing to shipping container security: can passive sensors bring a solution?

G. Janssens-Maenhout [a,*], F. De Roo [b], W. Janssens [c]

[a] Joint Research Centre Ispra, IES-CCU, Via Fermi 2749, TP. 290, I-21027 Ispra, Italy
[b] Ghent University, Faculty of Engineering, EESA, Belgium
[c] Joint Research Centre Ispra, IPSC-NUSAF, Italy

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ABSTRACT

Illicit trafficking of fissionable material in container cargoes is recognized as a potential weakness in Nuclear Security. Triggered by the attacks of 11 September 2001, measures were undertaken to enhance maritime security in extension to the Safety Of Life At Sea Convention and in line with the US Container Security Initiatives. Effective detection techniques are needed that allow the inspector to intercept illicit trafficking of nuclear weapons components or components of other nuclear explosive devices. Many security measures focus on active interrogation of the container content by X-ray scan, which might be extended with the newly developed tagged neutron inspection system. Both active interrogation techniques can, with the current huge volume of container traffic, only be applied to a limited number of selected containers. The question arises whether a passive detection technique can offer an alternative solution. This study investigates if containers equipped with a small passive detector will register during transport the neutron irradiation by fissionable material such as plutonium in a measurable way. In practice, 4/5 of the containers are about 1/8 filled with hydrogenous material and undergo a typical 2 months route. For this reference case, it was found that the most compatible passive detector would be an activation foil of iridium. Monte-Carlo simulations showed that for the reference case the activity of a 250 μm thin foil with 6 cm^2 cross-section would register 1.2 Bq when it is irradiated by a significant quantity of Reactor-Grade PuO_2. However this activity drops with almost two orders of magnitude for other fillings and other isotopic compositions and forms of the Pu-source. The procedure of selecting the target material for Pu detection is detailed with the theoretical methods, in order to be useful for other applications. Moreover the value of such additional passive sensors for securing maritime container transport is situated within the global framework of the First, Second and Third Line of Defense against illicit trafficking.

1. Scope of the illicit nuclear trafficking problem

1.1. From the heritage of the Cold War towards the possessive desire of extremist groups

The breakdown of the former Soviet Union in about 1991 raised an international nuclear security concern. Some nuclear material originating from the Soviet's civil-military coupled fuel cycle was present in three of the Newly Independent States (NIS) that had the status of Non-Nuclear Weapons States. The danger of illicit trafficking of nuclear material appeared real when in 1994 at the Munich airport, 363 g Pu was seized. (IAEA, 2007). The fear was that this was only a small fraction of the potential illicit nuclear trafficking. While the US focused on the removal of nuclear material or on physical protection, the EC transferred the knowledge for implementing a nuclear safeguards methodology with a state system for nuclear material accountancy and control.

In 1993 the International Atomic Energy Agency (IAEA) started to collect information on nuclear trafficking, which led to the Illicit Trafficking Data Base, the officially recognized reliable source for authoritative information on trafficking incidents. This Database now contains more than 1000 confirmed reports (with additionally 800 unconfirmed) on incidents involving smuggling, theft, loss and illegal disposal, illegal possession and transfer, and attempted illegal sales of the material. In 1995 the G-8 Moscow summit addressed a programme for preventing and combating illicit trafficking in nuclear materials and a corresponding international technical working group of international experts was established.

Figs. 1 and 2 show the distribution of nuclear illicit trafficking incidents over time per region and per type of material (more details

* Corresponding author. Tel.: +39 0332 78 5831; fax: +39 0332 78 5704.
E-mail address: greet.maenhout@jrc.it (G. Janssens-Maenhout).

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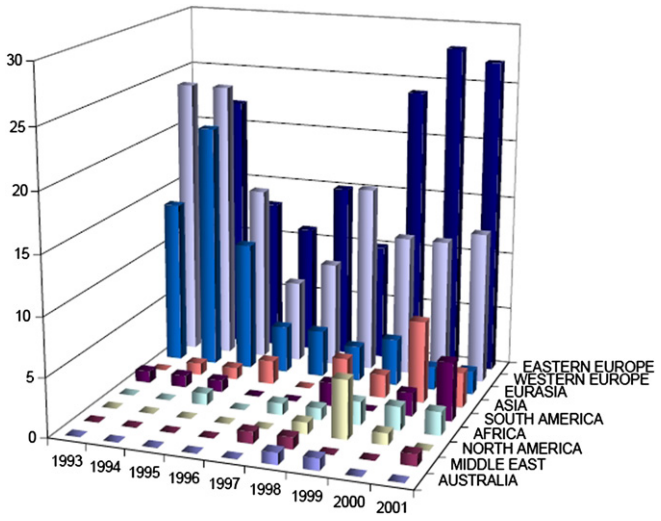

Fig. 1. Regional distribution of reported pre-9/11-era incidents.

are reported in (Bonny et al., 2005) and explanations of trends are proposed in Potter and Sokova (2002)). The trafficking of plutonium (Pu) and high enriched uranium (HEU) remained very low (< 5 per year) and reported cases with non-fissionable radioactive material increased.

With the attacks of 9/11, there was growing government and public concern that nuclear and other radioactive material may fall into the hands of non-state actors, such as terrorist groups or transnational crime networks. The A.Q. Khan Proliferation Network established a successful extensive black market for nuclear equipment, know-how and material, which was definitely flourishing on world-scale until 2003 (Janssens-Maenhout et al., 2007). Although the Khan Research Laboratories were originally only supplying their excess inventory of enrichment technology components to other countries (China and North Korea), a procurement network grew, which was later also partially supplying Iran and Libya. The sea routes between Pakistan, Dubai, and Malaysia were most frequently used for the shipment. For a number of cargoes to Iran and Libya, Dubai was a major transshipment point. It is now recognized that the clandestine network was possible: (1) partially due to the weakness in nuclear export controls even by members of the Nuclear Suppliers Group (NSG), and (2) partially due to the passive consent by some governments.

In the aftermath of 9/11 events, the G-8 Kananaskis summit fostered the World Customs Organisation (WCO) to concentrate efforts on enhancing the security of the global supply chain. In 2002, the WCO adopted under its SAFE framework the WCO Council Resolution, forming a Joint Customs-Industry Task Force on Security and Facilitation. Amendments to the existing Safety of Life at Sea Convention were adopted of which the most far-reaching enshrines the 2004 International Ship and Port Facility Security Code, with new maritime security measures for governments, port authorities and shipping companies (IMO, 2004).

But much more was done as described by Orlov (2004). The G-8 Evian summit of 2003 launched a Global Partnership Program against the Spread of Weapons and Materials of Mass Destruction (WMD), which was in line with the Proliferation Security Initiative of the U.S. In April 2004, the U.N. Security Council adopted Resolution 1540, requiring an effective implementation of measures to secure know-how, equipment and materials that could be used for WMD and to control export, and in April 2005 the International Convention for the Suppression of Acts of Nuclear Terrorism was adopted. The latter entered into force in July 2007.

1.2. The weak link of maritime container transport for a secure supply chain

Maritime container transport trade has intrinsically the potential to be the vector for security threats to a vast proportion of the coastal population. The detection of nuclear smuggling remains difficult because shipping is vital to the modern globalised economy. About 95% of the world's cargo moves by ship, corresponding to over 48 000 000 full cargo containers per year exchanged between major seaports. The size of the containers doubled from 20ft to 40ft (i.e. 12 m × 2.6 m × 2.6 m) and new vessels carry 8000teu[1]. In 2000, container traffic reached 100 000 000 teu, and it continues to increase about 8% per year according to Dahlman et al.(2005).

The shipping industry is vulnerable to criminal activities for several reasons (Flynn, 2004):

- The huge traffic volume leads to the fact that less than 2% of the containers are inspected at harbours.
- The transport chain is far from transparent: no single authority or industry has the full responsibility for security from beginning to end.
- During filling each container, all trust is with the shipper and after packing the bill of lading is rarely verified.
- Containers are not tampered while in custody in the harbour: transfer or re-packing and road transportation is often in the hands of a single person.
- Shipping companies are very reluctant to share information (which is of commercial value) with the harbours and customs authorities.
- The characteristics of the carrier vessels are cumbersome and far from standardized (most of the EU fleet is registered outside the EU).
- Container seals today are not difficult to remove and can be reproduced or forged.

This problem has been recognized by the OECD (2005) and the U.S. in particular. As such, the U.S. launched in 2002 the Container Security Initiative, focusing on screening containers at major European and Asian ports. Most emphasis is put on technological solutions for the security and logistic problems with the hope that technology could lead to a kind of smart container. In the meantime, active interrogation systems for detecting fissile material have been further evaluated, including neutron interrogation techniques, such as by EURITRACK with tagged neutrons (Carasco et al., 2008) or by Massachusetts with low energy neutrons (Johnson, 2006).

Today's inspection practices consist of either random container verification or verification because of incomplete documentation or intelligence information. The inspection means are mainly based on the weighing and video surveillance techniques. As an example, in Le Havre, a truck enters the harbour container terminal after it was automatically weighed and after pictures of the driver, vehicle plate and container number were recorded. At point of loading/unloading, the container might be selected for a 3D X-ray scan. The Le Havre Cyroscan system is applied to about a maximum 20 containers per day, which is 0.5% of all containers that pass through Le Havre. To leave the container terminal, only the documents of the truck are checked. (Dahlman et al., 2005)

[1] teu = 20ft equivalent units.

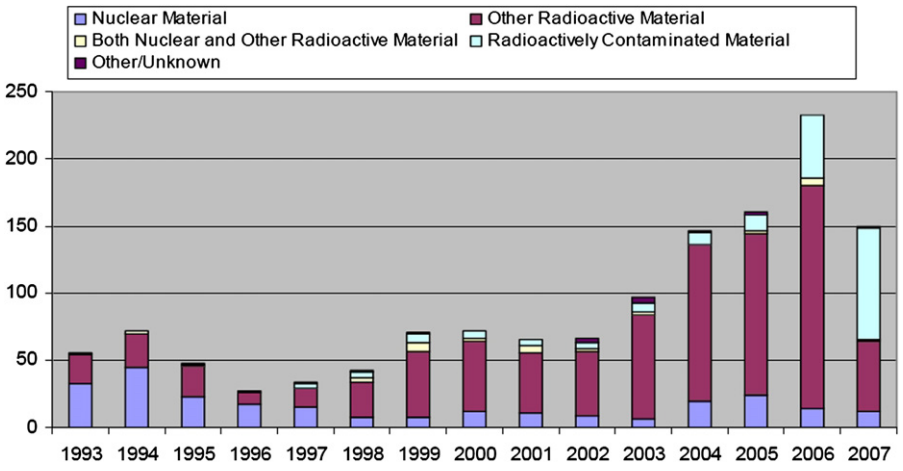

Fig. 2. Incidents confirmed to the ITDB as by Nilsson (2008).

2. Ways to combat nuclear illicit trafficking

2.1. The relevant nuclear materials

Under illicit trafficking of nuclear material, by definition, all nuclear materials are covered: this means both those from the civil cycle and those from the military cycle. To prevent from any loss, physical protection is combined with nuclear material accountancy and control for any type of nuclear material in both civil and military cycle in an appropriate manner[2]. In fact, when it comes to a real event of material interception/detection, the origin of the material in a first instance is irrelevant. It is clear however that, depending upon the character of the material, HEU and Pu deserve more attention than low enriched uranium, mixed oxide fuel or natural uranium. Whereas at the level of prevention, no difference needs to be made for civil versus military nuclear material, the same is not true with respect to the ability of tracing the potential origin/source. For civil nuclear material, good back-tracking can be done when at least a nuclear materials database is available. For nuclear material from military origin, it is expected to be much more difficult or even impossible to trace back to source.

As already envisaged in the Manhattan project, two paths are possible for constructing a nuclear device, either producing HEU, or producing Pu. At the time the Non-Proliferation Treaty came into force and Safeguards Agreements with Signatory Parties had to be drafted, the safeguards goals with significant quantities as shown in Table 1 were derived for each of these, based on critical mass estimations (e.g., Clark, 1980). To produce HEU, a physical isotope separation technique is needed, whereas the Pu production or breeding needs irradiation of fresh fuel in a neutron-flux and chemical reprocessing of the spent fuel. Even though the latter path seems longer, it might provide a quicker route because of the smaller amount of nuclear material and the higher accessibility to the reactor and reprocessing technology than for the tedious enrichment technology. History has demonstrated this preference at the nuclear tests by India in 1974, India and Pakistan in 1998, and North-Korea in 2006. This study contributes to a detection method of illicit trafficking of Pu, making use of the nuclear properties of ^{240}Pu (high spontaneous fission cross-section), and hence not of HEU trafficking.

[2] Even though it is expected that the physical protection for military material is significantly higher.

2.2. The legal framework to combat illicit nuclear trafficking

The legal definition of the acts committed during nuclear trafficking can be taken from the Conventions on the Physical Protection of Nuclear Material (CPPNM, 1980) (INFCIRC/274/Rev.1, 1980) and for the Suppression of Acts of Nuclear Terrorism (ICSANT, 2005) and include both (1) an unauthorized presence and unlawful possession and illegal disposal of nuclear material (CPPNM, Art. 7(a) or ICSANT, Art.2(a)), and (2) a violation of the regulations for obtaining, handling, and transport (CPPNM, Art. 7(b, c) or ICSANT, Art.2(b)).

These acts can be effective at three levels (Nilsson, 2008):

- The level of prevention: These acts are a consequence of a failure of physical protection at a certain location or an indication of undeclared nuclear material production and challenge the nuclear material accountancy and control, which should provide a First Line of Defense.
- The level of detection: While interrogation techniques at border control should reveal each significant quantity of nuclear material based on the radiation response, these interrogating measurements are exposed to radioprotection

Table 1
The three IAEA safeguards goals for nuclear material in the fuel cycle.

Safeguards Goal	Significant quantity	Timeliness	Probability[a]
^{235}U in Low enriched uranium	75 kg	1 *year*	$\alpha \leq 5\%$; $\beta < 5\%$
^{235}U in high enriched uranium	25 kg	4 *weeks*	$\alpha \leq 5\%$; $\beta < 5\%$
Natural uranium	10,000 kg	1 *year*	$\alpha \leq 5\%$; $\beta < 5\%$
Depleted uranium	20,000 kg	1 *year*	$\alpha \leq 5\%$; $\beta < 5\%$
Thorium	20000 kg	1 *year*	$\alpha \leq 5\%$; $\beta < 5\%$
Artificial uranium ^{233}U	8 kg	4 *weeks*	$\alpha \leq 5\%$; $\beta < 5\%$
Fuel-Grade/Reactor-Grade[b] plutonium	8 kg	4 *weeks*	$\alpha \leq 5\%$; $\beta < 5\%$
Weapon-Grade[c] plutonium	4 kg	1 *week*	$\alpha \leq 5\%$; $\beta < 5\%$
plutonium mixtures[d]	3 kg	1 *year*	$\alpha \leq 5\%$; $\beta < 5\%$
Pu in irradiated Fuel Assembly	1 Fuel Assembly	3 *month*	$\alpha \leq 5\%$; $\beta < 5\%$

[a] with probability of false-alarm α and probability of non-detection β.
[b] Fuel-Grade *Pu* : $7\%Pu\text{-mass} \leq {}^{240}Pu < 18\%$ *Pu*-mass; Reactor-Grade *Pu* : $^{240}Pu \geq 18\%$ *Pu*-mass.
[c] Weapon-Grade *Pu* contains mainly ^{239}Pu, little amount $^{240}Pu < 7\%Pu$-mass, as defined by (Pellaud, 2002).
[d] *Pu* isotopic compositions with $^{238}Pu > 80\%$ *Pu*-mass are excluded from the safeguards regulation.

Table 2
Comparison between passive detector types.

	Etched-track detectors	TLD	Activation foils	Neutron bubble detectors
Region	fast	thermal & fast	mainly thermal	fast (BD PND) thermal (BTD)
Threshold	100 keV	–	–	10*keV* (fast)
Lower detection	0.3 mSυ	σ-dependent	σ-dependent	1E-5 Sv (1-bubble; fast) 1-100.E-6 Sv (thermal)
Outreading measurement sensitivity	counting tracks dose/$E_{deposition}$ to (secondary) charged particles	glow curve to dose/ $E_{deposition}$ to (secondary) charged particles	activity flux to neutrons	counting bubbles dose/dose/ $E_{deposition}$ not sensitive to γ and e

measures. Efficient detection followed by an effective and safe handling at such seizure constitutes the Second Line of Defense.

- The level of response: These acts can indicate a wider potential smuggling network (financial or other), and can be used by non-state actors for a major disruption (e.g. Pu-contamination of water supply or large areas in major cities). The tracing back of the origin by nuclear forensics is needed to fight the root cause, as part of the Third Line of Defense.

The level of prevention is legally covered within the Safeguards Agreements and the Additional Protocol, in particular by the paragraphs on import-export control. The IAEA has reportedly called for an international framework for treaty-based export controls (Squassoni and Behrens, 2005). Through the Nuclear Security Plan for 2006–2009, the IAEA want to build on recent achievements in strengthening nuclear security by introducing an internationally accepted nuclear security framework of recommendations as soft law. In parallel, the NSG has considered different steps to improve the prevention including wider use of the catch-all provision. Catch-all clauses have been introduced by different countries but as weak links remain: (1) the different responsible authorities for the control of dual-use items, which are only partially in contact with the whole list of competent authorities in charge in the Member States of granting export authorizations, and (2) the description list of items, which is running behind the latest products of leading-edge research.

At the level of detection and response, the hard law of Annex I in the CPPNM and of Article 18 in the CSANT is strengthened by guidelines for the detection of and response to thefts and losses, unlawful possession and trafficking, illegal disposal and criminal and unauthorized use of nuclear and other radioactive material, which are disseminated through a new IAEA Nuclear Security Series of publications as part of IAEA's Nuclear Security Plan. Moreover this Plan provides for activities that include assessment and evaluation services, technical advice, human resource development programmes (including training for raising awareness for involved services) and, to a limited extent, deployment of needed technical equipment. For the latter, a Border Monitoring Working Group (BMWG) was created in 2005 under the auspices of the IAEA, with the US Department of Energy (US DoE/NNSA), with the European Commission (JRC, DG RELEX, DG AIDCO) and with the EU Council as members, to coordinate internationally the implementation of support programmes against illicit trafficking. Even joint training actions are included under the Second Line of Defense programme.

The Third Line of Defense is legally dealt with in a less effective way: Even though the IAEA has already in 1995 setup the Illicit Trafficking Data Base and a Nuclear Smuggling International Technical Working Group with Nuclear Forensic Laboratories, the reporting of incidents remains on a voluntary basis. A significant improvement is anticipated if the reporting would be required by law.

States increasingly recognize their responsibility to control unauthorized movement of nuclear and radioactive material and focus mainly on the Second Line of Defense. Efforts are made to secure national borders through the installation of radiation detection equipment and to ensure that frontline officers have adequate training and support to deal with and respond to seizures and detection alarms. Also the security of transport of nuclear and other radioactive material received more attention. A wide range of bilateral and multilateral initiatives to combat illicit trafficking are undertaken either following a so-called data-driven approach (looking for anomalities in large transport information datasets) or a process-driven one (verifying selected pieces of information registered at critical points in the transport process). Whereas the data-driven approach allows, with an intelligent information management system, to assess the vulnerability of the maritime transport chain, the process-driven one is expected to provide, with highly automated surveillance and monitoring techniques, the hard proof.

The US favoured surveillance and monitoring techniques and advocated even a 100% scanning of shipments, but the EU took a data-driven approach and fostered information exchange. A combination of both should guarantee continuity of knowledge, in particular of containers used in cargoes with suspicious routes. A contribution thereto could be given by applying for those cargoes the so-called "smart containers", which are equipped with embedded intelligent sensors registering the trail history.

Table 3
The ideal half-lift $T_{1/2ideal}$ in function of the decay time t_d and irradiation time t_c (in days).

$t_d \backslash t_c$	40 d	60 d	80 d
1 d	7.5 d	10.1 d	12.6d
2 d	9.1 d	12.1 d	14.9 d
3 d	10.4 d	13.7 d	16.7 d
4 d	11.6 d	15.0 d	18.2d
5 d	12.6 d	16.2 d	19.6 d

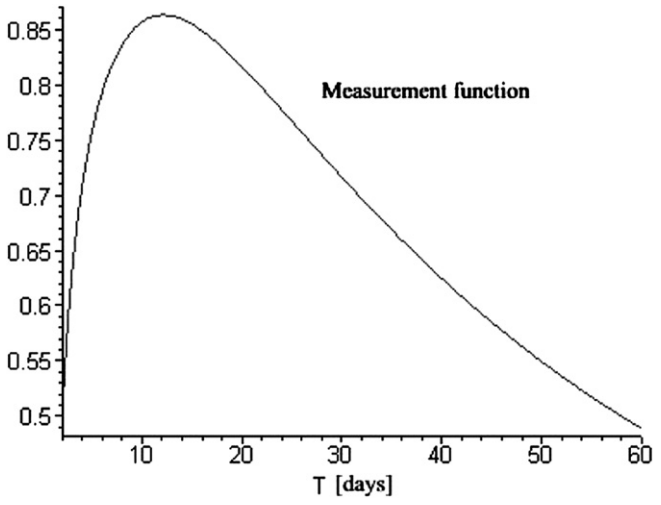

Fig. 3. Measured decay activity in function of half life.

Table 4
Ranking list of nuchde.s wrth most appropriate nuclear and physical properties.

Nuclide	Thermal capture	Av. fast capture	Weighed wiih $^{240}Pu_{sf}$	Daughter half-life	Abundance %	Molar volume cm^3
^{191}Ir	930*barn*	7.34*E5bam*	74.1*mbarn*	73.8 d	37.3	*8.52*
^{197}Au	98.7*barn*	2.99*E5bam*	30.2*mbarn*	2.7 d	100	10.21
^{185}Re	114*barn*	5.67*E5bam*	57.3*mbarn*	3.7 d	37.4	8.86

2.3. Raising alertness by equipping a container with a nuclear material sensor

Traditional sensors are electronic seals (radiofrequency identification, infrared, direct contact and very-long-range cellular or satellite seals), or intrusion detectors (mechanical, magnetic or electric switches, capacitance sensors, strain sensitive cables, taut wire sensors, ported coax buried line sensors, balanced buried pressure line sensors, photo electric beam sensors, microwave sensors, vibration sensors, fibre optic wire sensors, audio and acoustic detectors, passive and active ultrasonic sensors, geophone sensors, video motion detection sensors, radar), enlarged with smart dust sensors[3] (Kahn et al., 1999). Those sensors focus mainly on securing the container by verifying the integrity and the locking regardless the content shipped. To effectively fight against illicit trafficking of nuclear material in containers that are loaded in several steps at different harbours, a sensor that detects the presence of this nuclear material is needed.

2.4. Outline of the study

The investigations were performed in the framework of a larger feasibility study that focussed on a passive and small sensor and its detection capability for safeguarded fissionable material. The nuclear material for this study is Pu under different forms and different isotopic compositions. Given that ^{240}Pu emits neutrons because of spontaneous fission, it could be detected by passively registering the neutrons, if they reach the detector. The latter depends mainly on the material between the neutron-source and the detector and will determine the detection limits.

The choice for a passive detector implies a lower efficiency compared to an active detector, but the advantage of the passive detector is the much longer irradiation time (the passive detector can register the neutron flux during the total transport time). The key question is whether the detector efficiency of the small sensor allows it to differentiate the neutrons of the fissionable material from the background irradiation. The detection limits are determined by the detector type and size, its position versus the fissionable source and the neutron absorption of the potentially additional filling material in the container.

A series of neutron transport simulations were carried out to derive the neutron flux that can be measured from different types of Pu (reactor-grade and weapon-grade, in oxide and metal form) through different container fillings (air, hydrogenous[4] and steel). The irradiation of the detector was determined under the assumptions that the transport time between two successive harbours is typically about two months and that the delay between unloading of the content with potential Pu and the detector reading is some days. Typical cargo densities for steel[5] are below 0.5 g/cm^3 and for water in the range of 0.1–0.5 g/cm^3 (representing a container with food or wood). The stowage factor for cargoes of plastic, leatherwear, fruit is between 8.5 and 11.3 m^3/kg (Stopford, 1997). An average filling density of 0.12 g/cm^3 is in practice valid for at least 80% of the food containers and corresponds to a container that is for 1/8 of its height (29 cm) filled with a water layer. Such containers with 1/8 hydrogenous filling and shielding a bare metal sphere of 4 kg weapons grade Pu was chosen as a reference case. For this reference case the design of a passive sensor is addressed in the following Section 3, with simulations in paragraph 3.4 and experimental validation in paragraph 3.5.

3. Designing an appropriate passive sensor

3.1. Why an activation foil

Different types of passive detectors were evaluated, including: etched-track detectors, bubble detectors, activation foils and thermoluminescent dosimeters (TLD). Table 2 summarizes the literature (Knoll, 1989; Luszik-Bhadra et al., 1993; Tanaka and Furuta, 1976; Lakshmanan, 1982; Ing et al., 1997; Gambarini and Sinha Roy, 1997; Morelli et al., 1999; Ipe et al., 1992; Tanaka and Furuta, 1977) and compares the different passive detectors. It does not give a quantitative comparison on the detector efficiency because the efficiency of most detector types is investigated for specific doses or energy deposition. An evaluation of various activation targets (Au, Cu, Mn-Cu, In, Fe, Al, Ni, Mg, Ti, U) is typically done when using the activation foils to adjust the neutron spectrum in a reactor flux (e.g. the Material Test Reactor PARR-1, Malkawi and Ahmad (2002)). For the passive detector required for shipping containers, it is the flux (at very low values) and not the energy that is of interest.

For the purpose of this study, an activation foil is the most appropriate, because the measurement focuses on the neutron flux. The activation foil is simple to use, and it has small dimensions. After irradiation the measured activity of the foil is related to the mainly thermal and epithermal neutron flux to which it was exposed. In this way a large activity (sufficiently larger than the activity induced by natural background neutrons) can indicate the presence of an artificially increased number of neutrons, emitted also by spontaneous fission, such as from ^{240}Pu and by other neutron sources such as α-decays causing (α,n) reactions (Reilly et al., 1991).

3.2. Technical requirements due to container transport constraints

In order to appropriately select the material of the activation foil, the following characteristics were investigated:

- The neutron capture cross-section of the material for thermal or fast neutron flux, the latter being weighed by the Watt spectrum for ^{240}Pu, which should be high;
- The abundance of the isotope, which should also be high[6];
- The half-life of the decaying element, which should be approaching the ideal half-life T_{1/2ideal}.

[3] To be interpreted as autonomous micro-electromechanical systems.

[4] Hydrogenous medium is the strongly interacting medium in the simulations with water, the common componeant of many organic goods.

[5] These steel cargo densities are because of payload restrictions smaller then 1/15 of the nominal steel density of 7.8 g/cm^3.

[6] Nuclides with small abundance still remain interesting if enrichment is considered.

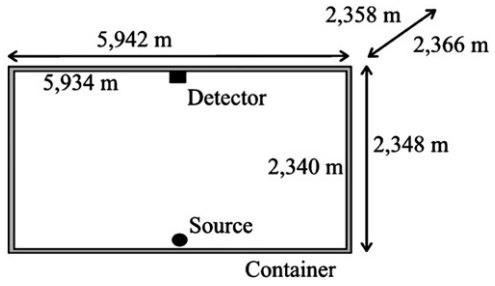

Fig. 4. Model of a 20 foot container.

This ideal half-life T_{1/2ideal} was derived mathematically under the assumptions that (1) the time dependence of the neutron flux Φ is negligible, (2) the burnup of the mother element (N1) with neutron capture cross-section σ is much smaller than the decay of the daughter element (N2) with decay constant λ and therefore a constant concentration of the stable mother element: $N1(t) = N1(t_0)$ referring to a starting point t_0. The activation of N1 into the decaying N2 is described by

$$((dN2)/(dt)) = \Phi\sigma N1 - \lambda N2,$$

with as solution:

$$N2(t) = ((\Phi\sigma N1(t_0))/\lambda)(1 - \exp(-\lambda(t - t_0))).$$

If $t_1 = t_o + t_c$ is the time that the flux is removed after an irradiation period t_c, then the activity of the daughter element is

$$A2(t) = \Phi\sigma N1(t_0)(1 - \exp(-\lambda t_c))\exp(-\lambda(t - t_1)).$$

Measuring from a time $t_1 + t_d$ until $t_1 + t_d + t_m$ after a decay period t_d and during a measurement period t_m yields the integrated decay activity:

$$M = \Phi\sigma N1(t_0)/\lambda(1 - \exp(-\lambda t_c))\exp(-\lambda t_d)(1 - \exp(-\lambda t_m)).$$

The T_{1/2ideal} is derived by differentiating M with respect to λ. The simplification that $t_m << T_{1/2ideal}$ allows to obtain the expression:

$$T_{1/2ideal} = t_c \ln 2/\ln((t_d + t_c/t_d))$$

The ideal half-life T_{1/2ideal} is tabulated as a function of the parameters $t_c = 60d \pm 20d$ and $t_d \in [1d,5d]$ and represented in Table 3. The corresponding decay activity which is expected for the measurement is shown in Fig. 3.

3.3. Selection of Iridium as target material

To find the most appropriate nuclides for the activation foil, all unstable nuclides with appropriate half-lives were listed. The parent that absorbed a neutron was traced back, and its capture cross-section looked up, using the Karlsruhe Nuklidkarte of 1998 and the NEA Databank JANIS 2.1 of 2004.[7] The nuclides were ranked, taking into account the natural abundance and molar volume, as explained in more detail by (De Roo, 2006).[8]

Iridium (^{191}Ir) was the most appropriate, followed by gold (^{197}Au) and rhenium (^{185}Re), as shown in Table 4.

Table 5
Normalisation factors for the Weapon-Grade (WG) and Reactor-Grade (RG) Pu-sources.

		WG Pu	RG Pu
Isotopic composition of Pu in mass fractions	Pu-238	0.012%	1–3%
	Pu-239	93.8%	60.3%
	Pu-240	5.8%	24.3%.
	Pu-241	0.35%	9.1%
	Pu-242	0.022%	5%
Total mass Pu-metal		4000 *g*	8000 *g*
Total mass PuO_2		4536 *g*	9069 *g*
n-activity for Pu-metal (sf)		2.4E5 *n/s*	2.9E6 *n/s*
n-activity for PuO_2 (sf+(α,*n*))		4.5E5 *n/s*	5.0E6 *n/s*

The isotope ^{191}Ir is very neutron-sensitive, because of the dominant (n,γ) reaction with a thermal capture of 930barn. Most of ^{191}Ir first transforms to ^{192}Ir^m but this product decays into the ground state of ^{192}Ir with a half-life of 1.44 min. The half-life of the activated ^{192}Ir is 73 days. As a consequence, the delay before measurement t_d has no significant impact. Natural Ir consists of 37.3% ^{191}Ir and of 62.7% ^{193}Ir, but the presence of ^{193}Ir can be neglected because of the lower neutron capture of ^{193}Ir and the short half-life of ^{194}Ir. The additional activity due to the irradiation of ^{193}Ir drops exponentially and is <10% after $t_d > 3d$. This activity is conservatively not taken into account but ameliorates considerably the detection capability of on-the-spot measurements with small t_d.

The choice of an activation foil of Ir implies to focus the analysis on the thermal and epithermal flux. In addition, the background flux is comparatively lower in the thermal region and thermal neutrons can be shielded more easily. As the activity of ^{192}Ir is anticipated to be very low, spectral information (γ/β) of ^{192}Ir is needed for the reading. For a fast and simple measurement, the 3 most intense γ-lines around 300 keV can be used. More extensive methods for measuring low activity, e.g. (Tarela et al., 1994) could be applied, but these go beyond the scope of this study on a "hand-held" detection device for on-site inspection of the container.

3.4. Simulation of the ^{191}Ir activation foil that is irradiated by ^{240}Pu

The simulations were carried out on a hypothetical 20ft bulk container (Fig. 4). The container had inner dimensions of 5.392 m × 2.358 m × 2.340 m, steel walls of 8-mm thick and a maximum payload of 18,500 kg (corresponding with a maximum filling density of 0.6 g/cm^3). The source was placed in the middle of the container floor and the detector was placed at the middle of the container roof. This geometry represents an average, because in a 20ft bulk container the root mean square(rms) distance between an arbitrarily placed detector and source is 2.29 m.[9]

Different types of filling of the container were considered, starting from a void container (with only air), and filling it gradually with hydrogenous content (food, wood, plastics, etc., modelled by water[10]) and a mixture of structural material and plastics (modelled by steel and water). The use of steel, air and water is not only a good choice for the usual content of a container, but is also very representative for different classes of neutron interaction. Specifically, air interacts weakly with neutrons, steel interacts

[7] Nuclides that capture a neutron and which decay into a chain out of which one of the daughters have an appropriate half-life are taken into account if all the foregoing nuclides have half-lives that are considerably shorter and if the nuclide immediately following in the chain is practically stable.

[8] For abundances different from 100% it was also verified if the other isotopes do not induce negative impacts.

[9] The chosen source-detector configuration with a distance of 2.348 m (the container height) approaches (with a slight conservative overestimation) the real average distance over the whole container volume. With $d^2 = x^2 + y^2 + (H - z)^2$ and assuming no favourite hiding place the root mean square (rms) distance is: $\text{rms} = \sqrt{\frac{1}{V}\int d^2(x,y,z)dxdydz} = 2.29\text{m}$.

[10] Water is reminiscent for organic material and plastics because the H-interaction is dominant for neutrons.

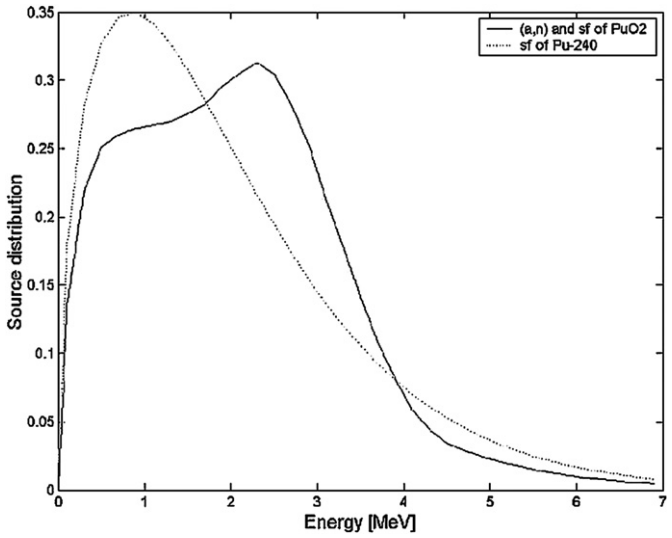

Fig. 5. Source distribution of PuO_2 compared to Pu.

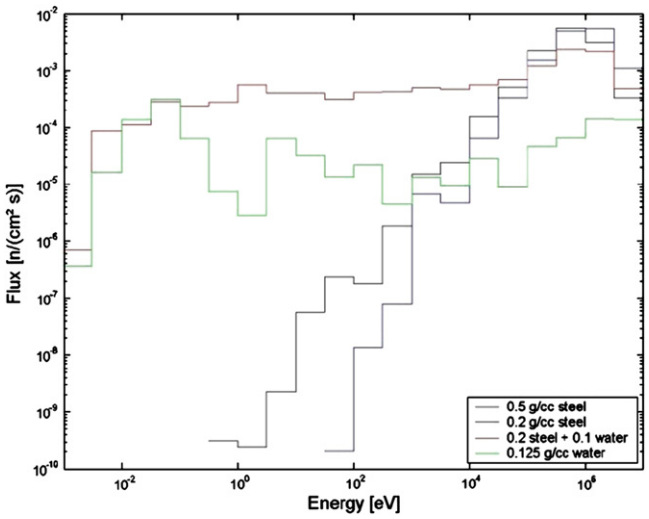

Fig. 6. Spontaneous fission-spectra of Pu-240 and cf-252.

intermediately (absorption) and water interacts strongly (both moderation and absorption).

The sources in the various simulations consisted of Weapon-Grade (WG) Pu metal and Reactor-Grade (RG) PuO_2. The neutron source was normalised with the mass fractions[11] of Table 5 and taking the sum of the spontaneous fission and (α,n) neutrons. Fig. 5 compares the source distribution of PuO_2 to Pu. The discussion of the sensor's feasibility focussed on the detectability of significant quantities of Pu, as defined in 2.1. The n-activity for WG Pu is one order of magnitude smaller than for RG Pu. Therefore 4 kg WG Pu-metal has been chosen as conservative reference case. This paper reports the most interesting cases around the conservative reference case. More is reported by De Roo (2006).

The differential neutron flux at the detector[12] is determined by the propagation of the spontaneous fission neutrons and the (α,n) neutrons out of the source to the detector. The content of the container will influence this propagation by moderating the source neutrons towards lower energies, and by attenuating the flux because of neutron absorption. It was verified that the method of filling can be neglected for the density ranges mentioned above: the Monte-Carlo simulations can for a small source and small detector not differentiate whether the material is stored on the container floor or distributed homogeneously throughout the container. The Variance Reduction Technique was applied for the homogeneous media under low flux with DXTRAN. (*MCNP*™, 2000)

All simulations demonstrated that the neutron flux at the detector depended linearly on the amount of source material. This is a consequence of taking the source as a homogeneous sphere and of the high density of Pu, implying small source radii. A container case filled with dry air is mainly governed by the geometrical attenuation, which can be described as the mathematical drop-off of the neutron flux propagating in three dimensions. The other cases are more governed by the moderation, and so represented by a double-peaked energy spectrum of the flux. For the hydrogenous content cases, the simulations showed for the analysed range of filling densities, a first peak around 1MeV that stems from the shape of ^{240}Pu spontaneous fission Watt spectrum and a second peak around 0.1 eV with a strength that depends on the amount of moderation. The mixed steel-water content case in Fig. 6 shows that for the neutron flux behaviour, a hydrogenous content is reminiscent but that the iron nuclei cause some more attenuation of the neutron flux for lower energies.

Table 6 confirms the very low values of the neutron fluxes on the detector, which have been calculated with a statistical relative error between 2.1% (for the lowest flux value) and 10.2% (for the largest flux value). These low flux values have to be confronted with the natural neutron flux background, that predominantly consists of cosmic neutrons and depends on the atmospheric height and geomagnetic coordinates.[13]

The neutron background rate is taken from different literature sources (Ziegler, 1998; Hess et al., 1959; Craig, 2007; Gordon et al., 2004; NCRP, 1987), which varied around 0.015 n/cm^2/s. Table 6 represents a conservative estimate for the neutron background flux at sea level with the maximum of the neutron flux restricted to latitudes common for containership trajectories (i.e. neglecting the poles). The background in the thermal energy range is almost 50 times smaller than in the fast one, and leads to the choice of the thermal energy range as operational domain for the activation foil. Even in the thermal energy range, the background remains 1.5–20 times larger than the neutron flux from the Pu-source. Therefore it was decided to introduce a cadmium layer on top of the Ir foil to shield the thermal background.

In practice, the latter is just an option because the giant dipole resonance causes an Ir capture just above the cadmium cut-off. Background neutrons with corresponding energies (about 0.65 ev) will have a non-negligible probability of being absorbed. This adds some uncertainty to the measurement for epithermal energies (which is not present at higher energies because of the significant decrease of the ^{191}Ir cross-section). Therefore the background uncertainty has to be determined by measuring separately the background (either the whole background without shielding or only the fast background when the foil is shielded) and this has to be subtracted from the total measurement.

To evaluate the activity of the foil after an irradiation time of $t_c = 60$d, the simulated neutron flux was convoluted with the differential cross-section of Ir. An exponential decay of the activity

[11] The exponential decrease of Pu was neglected because of the long half-life of Pu-isotopes compared to the 2 months container transport time.

[12] Average flux tallies "f4" were used for the detector cell.

[13] Other contributions to the neutron background which result from artificial neutron sources (e.g. inside a nuclear power plant) were neglected for this study.

Table 6
Integral flux values at the detector (target foil) over the energy range of 1 meV — 1 MeV.

Source	Filling	Integral flux		
		1meV – 1 eV (n/cm^2/s)	1 eV – 1keV (n/cm^2/s)	1*k*eV – 1MeV (n/cm^2/s)
4 kg WG Pu-metal	1/8 H_2O	7.8*E*-3	2.6*E*-3	1.3*E*-2
	1/4 H_2O	1.4*E*-3	2.1*E*-4	9.0*E*-4
	1/2 H_2O	6.6*E*-4	2.1*E*-4	2.8*E*-4
natural background[a]		0.01	0.2	0.5

[a] upper limits as of the National Institute of Standards & Technology, (36).

Table 7
Foil activities calculated for different combinations of sources. Weapon-Grade and Reactor-Grade plutomum, curium and californium, and for different precentages of hydrogenous filling.

Source	Filling	Ir-foil activity (Bq)
4 kg WG Pu under the form of Pu-metal	1/8 H_2O	3.3*E*-2
	1/4 H_2O	1.8*E*-3
	1/2 H_2O	1.0*E*-3
8 kg RG Pu under the form of PuO_2	1/8 H_2O	1.2
	1/4 H_2O	1.1*E*-2
100 g ^{244}Cm	1/4 H_2O	7.6
100 g ^{252}Cf	1/4 H_2O	1.8*E*6

during $t_d \approx 2$d was considered to verify that the contribution of ^{193}Ir can be omitted. Foil activities in the order of 1–1000 mBq were obtained and detailed in Table 7. A significant quantity of 8 kg RG Pu (9.069 kg PuO2) that is shielded by the hydrogenous fillings of the container with an averaged density of 0.12 g/cm^3 is expected to be detectable with a foil activity of 1.2 Bq. For a doubling of the hydrogenous material (0.25 g/cm^{3}density), this activity decreases to 11mBq and starts to approach the detection limit. Considering WG Pu instead of RG PuO2 implies an emitted neutron flux that is up to a factor 20 lower. Only cases of containers which are for about 1/8 filled with hydrogenous material will allow detection of a bare metal sphere of 4 kg WG Pu-metal with a considerable foil activity (33 mBq).

3.5. Benchmark with lab experiment

An experiment was set up with a real radioactive source in the PERLA laboratory of the JRC Ispra, in order to derive an experimental detection threshold. The simulated model was simplified[14] in the experimental setup by

- Taking a californium (Cf) source instead of a Pu-source,
- Using polyethylene as cargo, and
- Reduction of the container height.

The replacement of Pu by Cf is reasonable because both elements have a very similar spontaneous fission neutron energy spectrum, as shown in Fig. 7. The absolute differences of the spontaneous fission neutron emission were accounted for by renormalizing the flux. For the masses and elements in this study, the neutron flux in the detector was not influenced by the physical composition of the source and only depends on the shape of the emitted neutron spectrum and the container cargo. The behaviour of polyethylene towards neutrons is very reminiscent of water. The reduction of the container height was done because of practical considerations and was accounted for by compensating the distance with extra material attenuation in the simulations.

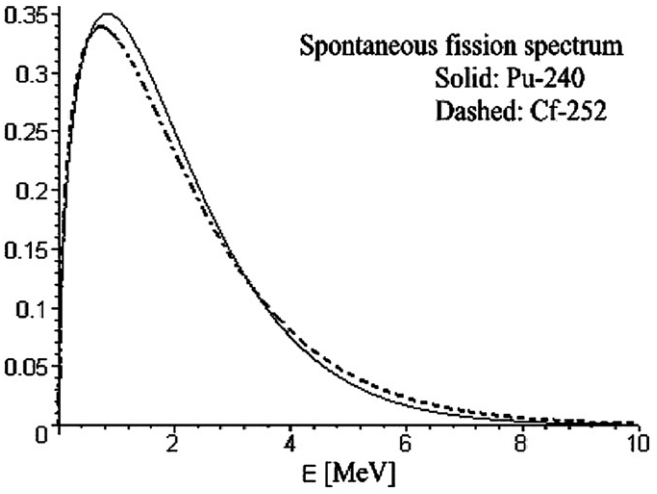

Fig. 7. Detector n-spectra for containers with 100 g WG Pu.

The experimental setup is sketched in Fig. 8 and dimensions are given in Table 8. The Cf source is placed on the table under the polyethylene blocks, which were covered with a Cd-layer. Three Ir foils have been used to measure the source. Between each foil there was 15 cm of polyethylene and the thinnest foil was placed at the lower position. The foils were irradiated for 48 days. The Cf source emitted 69,300 n/s at the start of the experiment down to 67,000 n/s at the end. Monte-Carlo simulation of experiment with geometry of Table 8 and with a constant[15] Cf as point source of 6.8 .10^4 n/s resulted in the histograms of Fig. 9, which mainly show the shadowing of the middle foil by the lower one and of the upper foil by the lower and middle one. Already these theoretical results show a very low flux, which suggest that only the lower foil will show an easily measurable activity. To determine the background neutron component in the experimental facility, an iridium foil of type "a" was kept separate for measuring the neutron background inside the experimental facility, but the latter (measured with the same detectors as the other foils) revealed no measurable activity. The paper further focuses on the evaluation of the lowest foil. The foil activation was measured by a Germanium γ-detector type GR 2020 over a large energy region. The results of the experiment are shown in Fig. 10 and summarised in Table 9.

Comparison between the experimental measurements of the foil activity and the Monte-Carlo simulations was made by conversion of the calculated gamma emission of the foil to a measured one. For this conversion, the γ-self-absorption of the foil (S = 0.98 for 0.125 mm Ir) was taken into account, as well as the detector efficiency and the relative peak intensities of Ir. The detector efficiency consists of a geometrical efficiency (8–mm distance between foil and detector, spherical angle $\approx 1.4\pi$) and an intrinsic efficiency. The latter is energy-dependent and was derived at PERLA (Favalli and Pedersen, 2006). Experimental internal conversion coefficients as recommended by (Baggerly et al., 1955) were applied.

[14] The scaling down by exchanging the source is commonly done at PERLA, e.g. (Tagziria et al, 2007), as well as at other locations by others, e.g. (Cucchiara et al, 1985).

[15] It was anticipated that the 1.7% improvement of results by taking into account the Cf decay can be neglected in the gross estimate (order of magnitude) of this study.

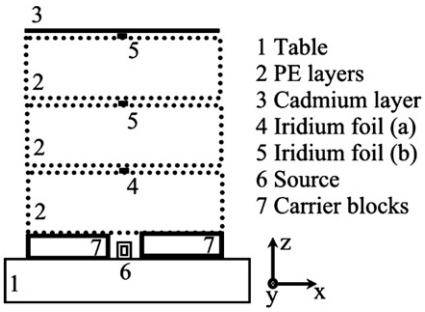

Fig. 8. Sketch of experiment (side view).

Table 8
Geometry of the experiment at laboratory scale.

	Dimensions		
	x (cm)	y (cm)	z (cm)
Polyethylene layer	50	50	15
Cd-layer	50	50	0.1
Thin Ir foil (type a in Fig. 8)	2.5	2.5	0.0125
Thin Ir foil (type b in Fig. 8)	2.5	2.5	0.0250

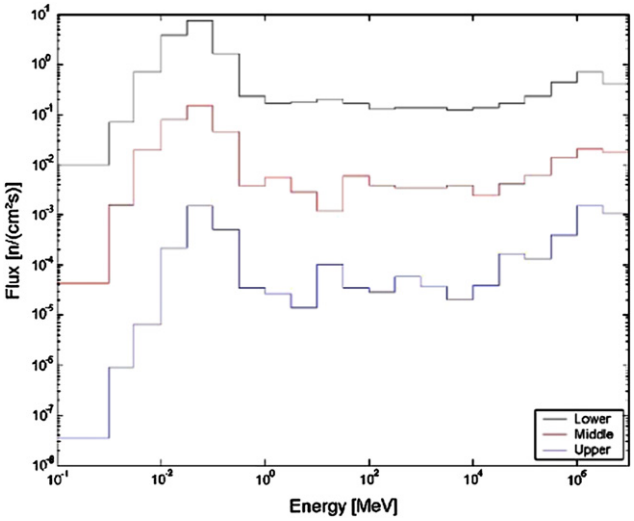

Fig. 9. Simulated n-irradiation in the 3 activation foils.

In Fig. 11 the calculated and measured gamma emission per second is compared for the three energy peaks of ^{192}Ir, which were selected based on the emission probability and on the gamma intensity. The experimental measured activity, on average 0.7 ± 0.1 Bq, was comparable with the averaged calculated activity of 0.75 Bq (simulations adapted to the experimental setup) for three different peaks of ^{192}Ir, with the most important peak at 316 keV. There was good agreement between the experiment and the simulations. However, the value for the most irradiated foil shows to be below 1 Bq, which complicates a reading of the activity on-site.

Iridium has several side peaks, which allowed concluding that the minimal measurable activity that could be distinguished out of the background was 10mBq for the germanium (Ge) detector used. A Ge detector is sensitive but has to be cooled and a measurement time period in the order of one hour has to be considered. This implies practical complications for an on-the-spot measurement of the foil. The detection capability can be potentially enhanced by applying very-low-activity detectors, which are tuned to the Ir-activity by adjusting the amplifier gain more to the 300 keV region and of which the response function is folded with the anticipated neutron spectrum of Ir. Today full-spectrum analyses of natural γ-spectra are measured with high-efficiency BGO scintillation detectors (Hendriks et al., 2001) and the high-sensitivity techniques (up to mBq) as used in radon detectors (Brunke et al., 2002) are available. However, such detectors are not standard equipment and their application therefore considered beyond the scope of this paper. More useful would be an enlargement of the dimensions of the foil ($25 \times 25 \times 0.25$ mm^3). The activity of the foil increases proportional with its cross-section and its thickness.

Of course, active detection techniques are much more precise in providing information on the container than a passive activation foil. Portal monitors at harbours are today equipped with 3D X-ray scan and might be extended with a active neutron interrogation techniques such as the tagged neutron inspection system

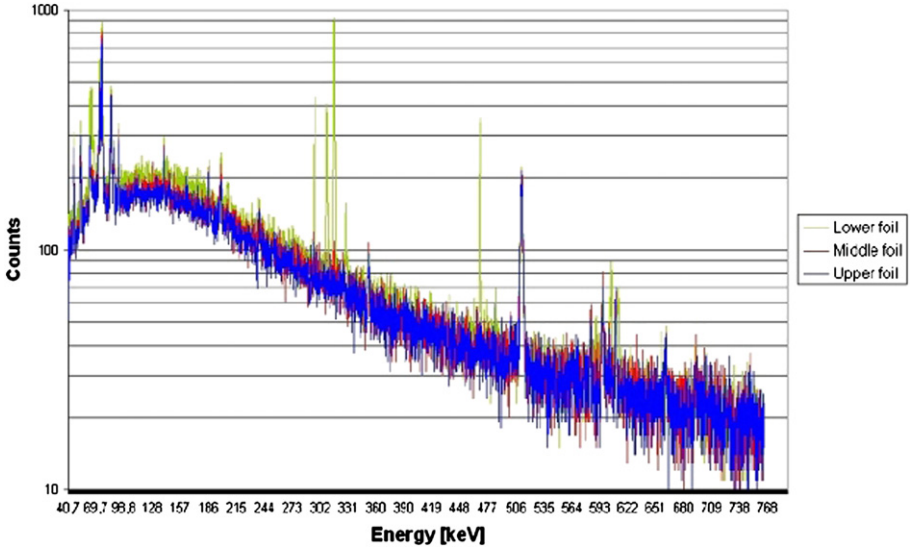

Fig. 10. Experiment (10–15 cps, dead tie < 1%).

Table 9
Measured gamma-emission of Ir- 192 in the lower foil (at position 4 in Fig. 8).

Energy (keV)	Rel. γ-intensity (%)	Counts (*cps*)	Error (±*cps*)	γ-emission (γ/*s*)	Activity (m Bq)	Error (m Bq)
295.95	36	2387	160	0.24	660	81
308.45	35	2029	159	0.20	571	82
316.50	100	6529	194	0.80	801	56

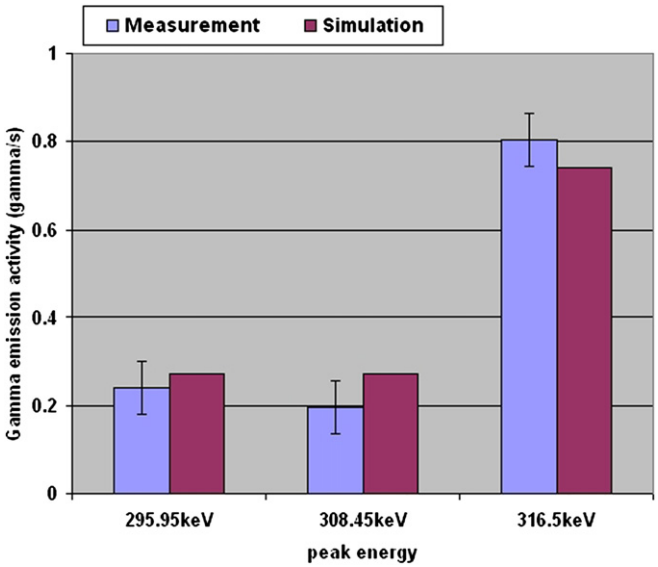

Fig. 11. Experiment and simulated activities for the detector.

developed in the EURITRACK project (Carasco et al., 2008). However active interrogation is only applicable to a very small portion of containers (less than 2% with today's container traffic). Moreover irradiation of containers with food might become complicated. The Food Additive Amendment provides that food is adulterated if it has been intentionally irradiated, unless the irradiation is carried out in conformity with a regulation on safe conditions of use (WHO, 1994).

4. Conclusion

To combat illicit nuclear trafficking in maritime container transport, a good level of detection is essential, and approached in a so-called Second Line of Defense programme with data-driven or process-driven technologies. Although the process-driven technologies are done now with a large range of surveillance and active interrogation techniques, passive sensors that register the radiation during the transportation route might be an interesting supplement because of its data-driven characteristics. In particular, for food containers, for which active scanning is strongly regulated if permitted, passive sensors might be an alternative solution to detect illicit nuclear trafficking. Based on information of suspicious cargoes and routes, the equipping of the most vulnerable containers with such sensors might not only enhance the detection capabilities, but also the level of prevention at the First Line of Defense. Moreover, they can contribute to strengthen the Third Line of Defense, if the information collected by the sensors during transport is internationally exchanged. Even though the reporting to the IAEA's Illicit Trafficking Database is (so far) on a voluntary basis, the need for international information exchange should be underlined, and ideally called for by law.

Containers filled with pallets of organics such as food, plastic, wood or leatherwear are more vulnerable for potential illicit trafficking of special fissionable material (in particular Pu) than bulk containers of ferrous materials and ores. The reasons are threefold: 1) their ordered structure allows easy access, 2) the surrounding hydrogenous layers are known as good shields for the neutrons, and 3) in the case of a food container, the active scanning is only permitted under special conditions. A practical detection system for Pu smuggling should be able to detect a significant quantity of Pu according to the IAEA safeguards goals. A feasibility study on the detection of 8 kg of Reactor-Grade Pu or 4 kg of Weapon-Grade Pu in a food container by a passive sensor was only partially satisfactory. The most suitable passive Pu detector for hydrogenous containers with a typical transport time of 2 months may be a small activation foil of Ir that is at one side covered by a Cd-layer (to shield for the thermal neutron background). Monte-Carlo simulations, validated with a benchmark experiment, demonstrated the detection constraints.

The Ir foil, fixed on top of the container will at inspection reveal the presence of 8 kg Reactor-Grade Pu (in the form PuO2) transported over 2 months, if the RG Pu is only shielded by hydrogenous container fillings of about 0.12 g/cm^3 density. In that case the corresponding neutron flux of 1.6 n/cm^2/s at the detector has induced after 2 months in the foil of 6.25 cm^2 and 0.25 mm thickness an activity of about 1.2 Bq, which is directly measurable on-site. Statistically, about 80% of the food containers have an average loading density between 0.1 and 0.2 g/cm^3 and for the higher loading factors the neutron absorption leads to lower activity. In the case of a doubling of the hydrogenous filling towards a density of 0.25 g/cm^3, the activity is reduced by two orders of magnitude to only 0.011 Bq.

To reveal by means of a passive detector the presence of 4 kg of Weapon-Grade Pu (in the form Pu-metal) is even more demanding. In the case that the 4 kg WG Pu-metal is only shielded by hydrogenous container fillings of about 0.12 g/cm^3, the induced foil activity mounts only to 0.033 Bq and for higher loading factors the activity drops beyond an easily measurable detection threshold (of 1 mBq). In the latter case, an upscaling is only possible by increasing the size of the foil. The activity of the foil varies proportionally with its dimensions, but taking thicker or larger foils will be more expensive and less discrete.

The choice of Ir is justified, in particular for a timely response on a thermal neutron flux. The combination of ^{191}Ir's half-life, neutron capture cross-section and abundance was most compatible with the technical requirements for a passive container detector; ^{197}Au was ranked second for this application.

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