Using Fast Neutrons to Detect Explosives and Illicit Materials

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Abstract. The present global political landscape continues to encourage the development of new technologies for the detection of hidden illicit materials, especially explosives, in packages ranging in size from small mail items to cargo containers. The various approaches using fast neutrons for contraband detection, presently under development, are reviewed.

1. Introduction

The bomb that brought down PanAm-103 shortly after take-off over Lockerbie, Scotland, on 21 December 1988, resulted in a massive political and scientific focus on the need to develop new explosive detection technologies [1, 2]. More recent terrorist attacks, especially those of 11 September 2001, have further spotlighted vulnerabilities and stimulated activities to prioritise the improvement of security associated with the aviation industry in particular.

Presently, the detection of concealed contraband is based mainly on the use of X-rays [3], vapour detection [4, 5] and sniffer dogs [6, 7]. X-rays are widely used for bulk package surveillance since they have many advantages [8]; the production and detection technologies are well advanced and relatively inexpensive, the machines are of a reasonable size and their presence is accepted in public places. However, X-rays suffer from the main disadvantage of having a small interaction probability with the low-electron density elements from which organic materials, including most explosives and illicit drugs, are composed. Therefore all these substances have undistinguished X-ray absorption or incoherent scattering characteristics. In addition, although some X-ray scanners can produce a sharp image as well as a density-dependent shading of the interrogated object, illicit drugs and explosives can be moulded or packed into any form. This makes their detection very difficult through shape recognition, the most commonly used method still used in X-ray inspections. There have, however, recently been a number of significant improvements in X-ray technologies, most notably the use of coherent scatter tomography [9-11].

The limitations of X-ray inspection techniques have stimulated the need to develop alternative methods, including those based on nuclear physics. This paper reviews the current status with respect to the use of neutron-based technologies for the elemental analysis of materials, in particular for the detection of hidden explosives and illicit drugs. Other reviews exist in the literature [12-19], and a number of papers have also recently appeared which deal with the use of nuclear techniques for the particular application of humanitarian demining [21-23].

2. Elemental composition of explosives and illicit drugs

Although there are many basic types of explosives and illicit drugs which can be combined and diluted to make hundreds of variations, most consist almost exclusively of the elements H, C, N and O. Furthermore, these substances are fortunately well-separated from most common

materials in one or more elemental features. Fig. 1 shows a stacked bar graph of the fraction of each constituent atom, as a percentage, for a selection of explosives, illicit drugs and miscellaneous materials. It can be seen that explosives are distinguished by relatively high proportions of nitrogen and oxygen and relatively low proportions of carbon and hydrogen. On the other hand, illicit drugs are generally rich in hydrogen and carbon and poor in nitrogen and oxygen. In addition, most explosives have densities in the range 1.2 to 2.0 g cm^{-3} which is generally larger than most everyday HCNO substances. These features may be utilized to identify the presence of explosives and illicit drugs hidden amongst the other material inside a closed container.

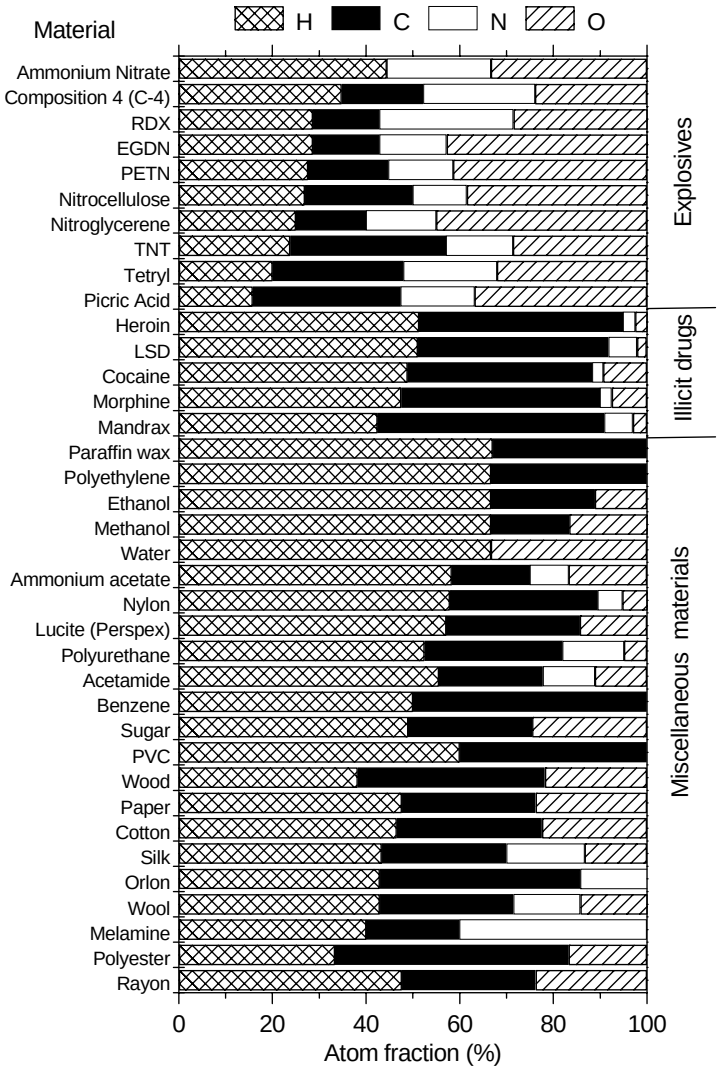

FIG. 1. Atomic fractions (as a percentage) of the elements H, C, N and O, which constitute a selection of explosives, illicit drugs and miscellaneous everyday materials.

3. Neutron-based techniques

Neutron-based techniques for the non-destructive elemental characterisation of materials in bulk have been in commercial operation since around the late 1970s, especially in the context of online analysis and control [24], most notably in the minerals industry [25]. Over the last decade there has been much interest in exploring the use of these techniques for the non-intrusive detection of explosives and illicit drugs, hidden in small mail items, airline baggage and large cargo containers [26]. Neutrons are effective probes for the elemental characterisation of bulk samples since they are not affected by electromagnetic forces and can therefore penetrate deeply into matter, interacting only with nuclei. When a neutron does interact with a nuclide, it is affected in a unique way that depends on the incident energy of the neutron and the species of the target nuclide. Most nuclides exhibit distinctive structure in their scattering cross sections which enhances the probability of certain neutron interactions occurring at specific incident neutron energies and scattering angles. In particular, the principal elemental constituents of narcotic and explosive substances (H, C, N and O) differ strongly from one-another in their interactions with neutrons and can thus be characterised via these differences. Neutron interrogation techniques generally rely on bombarding the nuclei in the interrogated object with neutrons of particular energy or energies, causing them to emit characteristic γ-rays or alter the energy of the interrogating neutrons. The attributes of the probing neutrons and the energy and spatial distributions of the detected radiation (neutrons or de-excitation γ-rays) may be used to determine the types, amounts and positions of the specific interacting nuclides, hence characterising the specific materials making up the interrogated object.

All nuclear inspection techniques are based upon specific neutron or γ-ray “signatures” which depend on the particular nuclear process being used. In most practical situations background components are significant, hence the sensitivity and specificity of the particular approach generally hinges on its ability to distinguish the signature of the concealed contraband substance clearly from the background. Therefore the scanning of the object might be necessary in order to distinguish, with high selectivity, between the signature of the contraband and the signals from the overwhelming majority of innocuous substances surrounding it. In such situations it is often advantageous to either nanosecond-pulse or tag the incident neutron beam.

It is possible to identify seven general neutron physics-based approaches which are being developed, each of which is briefly described below, and summarised in Table 1, where the acronyms refer to the operational names of the various techniques. There have been over 50 patents granted in this area in the United States since 1990, in many cases before displaying practical accomplishment. Furthermore, since there are commercial aspects to the development of many of these technologies, there has generally been inadequate reporting in the scientific literature. Although there has recently been a significant development of the use of beams of γ-rays, most notably nuclear resonance absorption [27] and gamma-ray transmission spectroscopy [28] , only techniques incorporating the use of fast neutron beams are discussed here.

TABLE 1: SELECTED FEATURES OF THE MAIN NUCLEAR PHYSICS-BASED TECHNIQUES FOR THE NON-INTRUSIVE INTERROGATION OF BULK SAMPLES.

Technique (see text)	**Radiation source ***	**Probing radiation**	**Main reaction type**	**Detected radiation**	**Primary (secondary) signatures**
TNA	^{252}Cf ; d-D or d-T STNG	Thermal neutrons	(n, γ)	Prompt γ-rays from neutron capture	H, N, C1 (others)
FNA	d-D or d-T STNG	Fast neutrons	(n, n'γ)	γ-rays from inelastic neutron scattering	C, O, Cl (N, others)
PFNA	ns-pulsed accelerator	Fast neutrons	(n, n'γ)	γ-rays from inelastic neutron scattering	C, O, C1 (N, others)
API	associated particle d-T STNG	14 MeV neutrons with associated α particles	(n, n'γ)	γ-rays in coincidence with α-particle	C, N, O (others)
PFTNA	μs-pulsed d-T STNG	Fast neutrons during pulse, and then thermal neutrons.	(n, n'γ) + (n, γ)	γ-rays from inelastic neutron scattering, capture and activation analysis	H, C, N, O (others)
PFNTS	ns-pulsed accelerator	White spectrum of fast neutrons	All available	Source neutrons which are transmitted	H, C, N, O, others.
FNGR	d-T STNG and ^{60}Co source	14 MeV neutrons and ^{60}Co γ-rays	All available	Source neutrons and γ-rays which are transmitted	H, C, N, O, others.
FNSA	ns-pulsed or DC accel.; STNG	Monoenergetic fast neutrons	(n, n) + (n, n')	Elastically and inelastically scattered neutrons	H, C, N, O (others)

* STNG: Sealed tube neutron generator

3.1 Thermal neutron capture and fast neutron inelastic scattering γ-ray signatures

(a) Thermal Neutron Analysis (TNA)

Fast neutrons produced by a radio-isotopic source such as ^{252}Cf, or a sealed tube neutron generator, are moderated to low energies (<0.025 eV) within the object under interrogation. A fraction of these thermal neutrons react with the nuclides in the object and the prompt capture γ-rays are detected [29, 30]. The application of TNA in the discovery of explosives is primarily based on the identification of nitrogen and hydrogen via the detection of the 2.22 MeV and 10.83 MeV capture γ-rays from ^{1}H from ^{14}N respectively. The capture γ-ray spectra measured in arrays of detectors may be tomographically analyzed to provide the density distribution of nitrogen and hydrogen within the package.

(b) Fast Neutron Analysis (FNA)

A collimated beam of continuous fast neutrons, typically from a sealed tube neutron generator, bombard the object under inspection. The de-excitation γ-rays released from nuclei activated in fast neutron inelastic scattering events are detected by an array of detectors surrounding the object and shielded from the direct exposure to source neutrons [31]. Specific elements present in

each volume element or "voxel" of the object are identified through the de-convolution of the γ-ray spectra measured by each detector [32]. The main signatures used are derived from detecting the 4.43 MeV γ-ray from ^{12}C, the 1.64, 2.31 and 5.11 MeV γ-rays from ^{14}N, and the 6.130 MeV γ-ray from ^{16}O [33]. Although the attenuation of fast neutrons within an object is much less than that of thermal neutrons, FNA imaging is also limited to smaller objects due to the lack of geometrical definition when there are large distances between inner voxels and the detectors.

(c) Pulsed Fast Neutron Analysis (PFNA)

The two limitations affecting FNA, namely the high backgrounds and relatively poor imaging capabilities for large objects, may be addressed by pulsing the incident neutrons and making use of neutron time-of-flight. An accelerator, such as a Van de Graaff, is used to produce nanosecond bunches of neutrons of energy around 8 MeV via a (d,n) or (p,n) reaction using a beryllium or lithium target. The neutrons are collimated and caused to scan the container under investigation and the γ-rays produced in inelastic scattering events are detected in arrays of NaI(Tl) crystals. The location of the interacting nuclide may then determined by measuring the time between the creation of the neutron in the target and the detection of the de-excitation γ-ray, thereby facilitating 3-dimensional elemental imaging [34, 35].

(d) Associated Particle Imaging (API)

The high cost and maintenance of a nanosecond-pulsed accelerator-based system is a serious drawback for most contexts. There are a growing number of manufacturers of sealed tube neutron generators (STNGs) which have the advantage that they are both compact and of relatively low cost, and produce intense continuous or μs-pulsed neutron beams. They utilize either the ^{2}H(d,n)^{3}He reaction (d-D) to produce 2.5 MeV neutrons or the ^{3}H(d,n)^{4}He reaction (d-T) to produce 14.1 MeV neutrons, and have found particular use in oil well logging, nuclear waste assay and neutron radiography [36]. STNGs are typically capable of producing 10^{8} to 10^{9} neutrons per second, some models as much as 10^{11} neutrons per second, and can be used in any orientation. Neutron generators incorporating the associated particle method have been developed in which the neutrons are tagged in time and direction by detecting the alpha particles which are simultaneously released in the opposite direction [37]. In API, gamma rays produced by inelastic scattering events are detected in a similar fashion to FNA and PFNA, although knowledge of the direction of the neutron and the time of its creation allows the position of the scattering nuclide to be determined, without the interrogating neutron beam being ns-pulsed.

(e) Pulsed Fast-Thermal Neutron Analysis (PFTNA)

The PFTNA technique generally utilizes a μs-pulsed sealed tube neutron generator which produces a train of 14.1 MeV neutron pulses, a few microseconds wide via the ^{3}H(d,n)^{4}He reaction. Fast neutrons incident upon the object initiate inelastic scattering interactions with elements including C and O. The de-excitation γ-rays are detected, and the events stored via a gated ADC. The neutron beam is then turned off for about 100 μs during which time a fraction of the fast neutrons remaining within the sample thermalise and are captured by elements such as H, N, Cl and Fe. The prompt capture γ-rays are detected by the same detectors and the events stored separately from the inelastic scattering spectrum. This procedure is repeated with a frequency of approximately 10 kHz. Every few hundred pulses, the neutron beam is turned off for a longer time interval, such as 3 ms, and a different ADC collects the events associated with the delayed

emission of γ-rays from the de-activation of elements such as O, Si, F and P. Therefore, by combining fast inelastic neutron scattering, thermal neutron capture and delayed activation analysis, a large number of elements contained in an object can be measured in a continuous mode without sampling [38, 39].

3.2 Fast neutron radiography signatures

(a) Pulsed Fast Neutron Transmission Spectroscopy (PFNTS)
The total scattering cross sections for fast neutrons exhibit resonance structures which are unique to the particular scattering nuclide. The PFNTS technique requires a ns-pulsed accelerator providing an intense "white" spectrum of neutrons produced via a (p,n) or (d,n) reaction on a thick target of Be or Li. A fan beam of neutrons (energies ranging from about 0.5 MeV to 10 MeV) is directed through the object under interrogation and the transmitted neutrons are detected in a bank of fast neutron detectors. Overley [40, 41] was the first to show that knowledge of the total cross sections for H, C, N, O, and other critical elements, allows the measured transmission spectra to be unfolded to provide aerial densities of elements present in the interrogated object. Tomographic imaging is also possible if the object is scanned from different directions [42]. Since the neutron transmission though the interrogated object is affected by small angle scattering, libraries of effective neutron transmission cross sections are needed for the specific geometry being used, either by direct measurement or sophisticated calculations that take account of the small angle scattering.

(b) Fast Neutron and Gamma Radiography (FNGR)
Simultaneous measurements of the attenuation of beams of fast neutrons and gamma rays may be used to produce a 2-dimensional projection image of the object under interrogation which may provide information on both the object's areal density and material composition [43]. The ratio R of the neutron and γ-ray attenuation coefficients can be determined (to a first approximation) directly from the measured neutron and γ-ray transmissions without knowing the mass of material in the radiation beam paths. The dependence on R on elemental composition, together with the imaging capability of the technique, can be exploited to provide information on the type and nature of the materials under scrutiny [44].

3.3 Fast neutron scattering signatures

(a) Fast Neutron Scattering Analysis (FNSA)
The neutron techniques described above are based on measurements of the attenuation of a pulsed fast neutron beam as it passes through the object being interrogated, or on measurements of characteristic gamma rays which are excited either by thermal neutron capture or by inelastic neutron scattering in the interrogated object. Fast Neutron Scattering Analysis (FNSA) is an alternative approach in which neutrons *scattered* out of the interrogated material are detected. The type, amount and positions of the nuclides responsible for the scattering are determined from measurements of the dependence of scattered neutron intensity and energy on scattering angle and the incident neutron energy [45, 46]. The FNSA technique consists of bombarding the sample of material being examined with a beam of monoenergetic neutrons, and using detectors to observe elastically and inelastically scattered neutrons at forward and backward angles. The incident neutron energy may be alternated between two carefully chosen values and neutron time-

of-flight can also be used if available, although is not mandatory. Measurements from all detectors are combined to form "scattering signatures" which are strongly characteristic of the species of scattering nuclide. It has been shown that FNSA can measure the atom fractions of the elements in a small sample (0.2 - 0.8 kg) of an HCNO material to an accuracy of a few percent and that explosive or illicit drug materials can be reliably identified from these measurements [47]. An important feature of FNSA is that it measures the elements which are essential for contraband detection (H, C, N and O) with similar sensitivity.

4. Recent developments of neutron-based systems

The non-intrusive screening of airline baggage for hidden explosives is by far the most difficult problem to tackle since this application has the most stringent requirements to satisfy in order to be both effective and acceptable [48]. Failures to detect explosives (false negatives) cannot be tolerated and the probability of incorrectly identifying innocuous objects as explosives (false positives) must also be small. Thermal Neutron Analysis (TNA) was the first neutron-based technique to be seriously considered for the detection of hidden explosives at airports. TNA systems, coupled to high resolution X-ray machines, have been intensively tested at a number of airports, including John F. Kennedy International Airport in New York, and Gatwick International Airport, near London [49]. TNA spectra, however, need to be corrected for substantial backgrounds resulting from the capture of thermal neutrons in the system shielding, detectors and other miscellaneous objects. Furthermore, spectral interference from competing elemental signals, low detector efficiency and serious signal attenuation resulted in the need for long measuring times for reliable screening of suitcase-sized objects. More recently, TNA systems have been marketed (by Ancore Corporation, and others) for the inspection of objects ranging in size from small parcels to motor vehicles for concealed bombs [50, 51], and seem to offer high reliability in situations where the interrogation measurement is not subject to undue time pressure.

The two approaches which rely on ns-pulsed neutron beams, viz. Pulsed Fast Neutron Analysis (PFNA) and Pulsed Fast Neutron Transmission Spectroscopy (PFNTS), have also been considered for use in an airport environment. Full prototype PFNTS systems for airline luggage screening have been developed and have undergone significant laboratory testing at the University of Oregon [52, 53] and Tensor Technology [54], and extensive Monte Carlo studies were completed at Argonne National Laboratory [55-57]. A recent review of a PFNTS system [58] by the (United States) National Materials Advisory Board concluded that PFNTS explosives detection devices are not ready for airport deployment for a number of reasons, including difficulties around cost, size, weight, and safety issues, and that PFNTS had not yet met the FAA's detection and false alarm certification requirements.

Neutron techniques that were originally considered for the primary scanning of airline luggage for explosives have recently found practical use in other, less demanding, contexts, which generally tolerate either a longer measuring time or a significant number of false positives and negatives. For example, PFNA systems designed by Science Applications International Corporation, and more recently Ancore, are being marketed for the elemental imaging of cargo containers at border posts [59] and are capable of screening five trucks per hour [60]. More recently, PFNA systems have been further enhanced by the inclusion of neutron and γ-ray transmission spectroscopy [61].

The PFTNA technique has been extensively developed into a reasonably compact system based on a μs-pulsed d-T STNG and BGO detectors, and marketed as PELAN (Pulsed Elemental Analysis with Neutrons). PELAN systems have been designed for the detection and characterisation of explosives and military ordinance [62] he dismantling of nuclear weapons [39] the detection of buried landmines. Further optimization of the signal-to-noise ratios of PELAN systems is presently being explored [63, 64]. Associated particle STNGs are also finding use in an number of contexts including the verification of chemical and nuclear weapons [37] and the detection of explosives and landmines [65].

A fully working prototype system making use of the FNGR technique has recently been built and tested by the group at CSIRO Minerals, Sydney [43]. The facility uses a 14 MeV STNG in combination with a 0.8 GBq ^{60}Co γ-ray source, and detection of the transmitted fast neutrons and γ-rays is achieved using banks of orange plastic and CsI(Tl) scintillators, respectively. High resolution false-colour images of fully-laden Unit Load Devices (ULDs) were obtained [44] which provided *R*-dependent information which could be used to distinguish between materials of different *Z*. A commercial-scale scanner is presently under development for in situ use at Brisbane airport by the end of 2005 which will achieve scanning and imaging times of about 1-2 minutes per ULD.

In addition to TNA as described above, there are a number of techniques which make use of neutrons from radio-isotopic sources such as ^{252}Cf, Am-Be or Pu-Be. The main advantage is the relatively low cost and long life of such sources, although the neutrons may not be "turned off." Thermal neutron capture using ^{252}Cf sources has been successfully applied to the identification of chemical warfare agents and high explosive munitions, and to the characterisation of unexploded ordnance from firing ranges and former defence sites [66]. One form of this technology has been developed commercially as PINS (Portable Isotopic Neutron Spectroscopy) Chemical Assay System and has been used by the United States army since 1992 [67]. It has also been shown [68, 69] that fast neutron transmission and scattering measurements using a ^{252}Cf source may be used to identify explosive-like materials. More recently, it has been shown that HNCO materials can be identified by detecting and analyzing elastically backscattered neutrons from a Pu-Be source [70, 71]. Although most of these approaches make use of the full spectrum of neutrons produced by the source, Kuznetsov [65] has reported on the use of a ^{252}Cf source in which the detection of the recoiling fission fragments allows ns-timed measurements to be made.

5. Possible future application of neutron methods to the interrogation of airline baggage

It is not clear at present whether any of the new neutron technologies will satisfy the stringent requirements of high throughput and low false identification rates that are demanded for the primary screening of airport baggage [72]. X-ray-based methods for detecting explosives are already accepted and widely used in public environments such as airports, and can satisfy safety requirements such as radiation shielding without undue difficulty. Neutron-based technologies face much greater problems in satisfying safety requirements due to the special radiation hazards particularly associated with neutrons, and because the shielding of intense sources of neutrons is more difficult and requires more space and material than is required for X-ray sources. These considerations together with an estimated measuring time of 6 seconds per item imply that

neutron-based screening systems are unlikely to be considered as direct replacements in the near future for the X-ray screening units now commonly in use at airports. However, for certain explosives the minimum quantity of material representing a threat may be as small as 0.2 kg. Therefore X-ray systems which attain the required low rate of false negative identifications might do so at the price of a false positive rate of several percent of the packages screened. This could lead to serious delays and other problems in operations with these systems, assuming that manual inspection of packages or baggage identified as suspect would then be required.

Neutron-based systems therefore might find use as the second (or third) stage of a multi-stage screening system for the detection of explosives in departing airline baggage. The initial screenings in such a system would use other methods which are capable of screening packages rapidly, such as X-ray imaging and odour sensing, in combination with any other *a priori* data resulting from passenger profiling and specific intelligence [73]. Packages identified as suspect by this system would be carefully diverted for a second screening by the neutron system, which would be required to confirm or reject the assignment made in the first screening. The number of packages to be processed in the second stage should be much smaller than in the first stage, assuming a false positive rate of less than 2% of the packages screened in the first stage. Therefore the slower throughput expected from the neutron system should not be a problem and a single neutron system should be able to process the work diverted to it by several first-stage screening installations.

A proposed neutron-based system for the detection of explosives or illicit drugs in luggage is shown schematically in Fig. 2. A continuous beam of monoenergetic neutrons, produced by a sealed tube neutron generator or other small, relatively cheap accelerator such as a radiofrequency quadrupole linac, is fan-collimated through the luggage conveyor belt. A γ-ray source such as ^{60}Co may be used as well. Neutron and gamma-ray detectors are situated at zero degrees and other forward and backward angles, and multiplexing the energy of the neutron beam though two or three specifically chosen values may be advantageous. The signature for an explosive (or illicit drug) would be derived from a combination of neutron and gamma-ray signals which would be processed slice-by-slice of the passing suitcase. Is it curious to note that most of the neutron-based technologies described above have made use of only one or two of the signatures listed in Table 1, probably as a consequence of the commercial aspect to their development. However, by detecting both the transmitted and scattered neutrons (and possibly the transmitted γ-rays), in coincidence with the de-excitation γ-rays produced in the object, the primary signatures of TNA, FNA, FNSA, PFTNA and PFNTS would be fused, thereby maximizing both the flexibility and reliability of such a system.

A system such as that shown in Fig. 2 would also be useful in the detection of illicit materials in incoming luggage. The requirements specified for detecting illicit drugs are much less stringent than those for explosives and a system capable of detecting a minimum quantity of 0.5 kg of illicit drugs in typical airline baggage with an efficiency of 90% would be extremely useful by customs or other authorities responsible for detecting contraband. Furthermore, since checks for illicit drugs are usually more focused on incoming baggage than departing baggage, occasional delays should be acceptable if extra time is required to resolve doubtful cases.

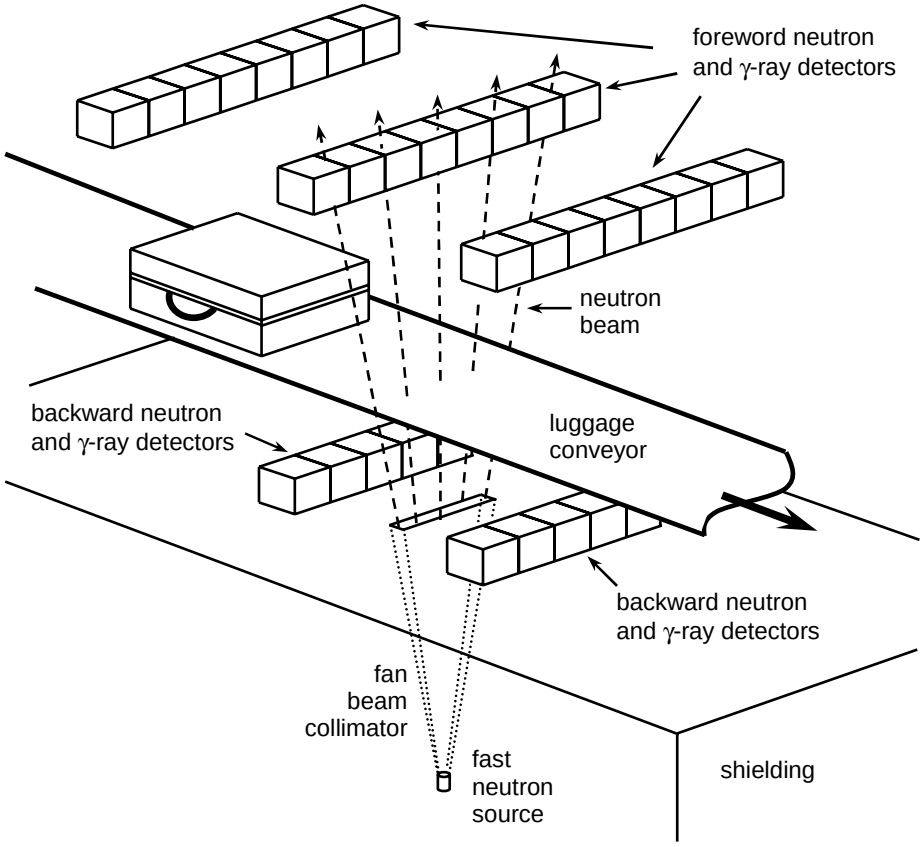

FIG. 2. A proposed neutron-based system for screening airline luggage. A beam of mono-energetic neutrons is fan-collimated through the baggage conveyor belt in a well-shielded vault. Neutrons and gamma rays are detected in arrays of detectors at backward and forward angles.

6. Conclusion

Over the last decade, technologies using fast neutrons have found use in a variety of contexts from the elemental characterisation of munitions, to the imaging of shipping containers. However, with the current state of the art, there is not compelling evidence that fast neutrons offer a practical alternative to X-rays as the primary probes for the detection of explosives in airline luggage. Future results from laboratory testing of prototype fast neutron-based systems, coupled with the necessary evolution of security policy and protocols, might result in neutron systems forming part of a multi-stage screening process in airports. It is more likely in the near future, however, that fast neutrons will find increasing use in the fight against the smuggling of illicit goods in air and shipping cargo, and in the search for buried landmines.

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