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ASSOCIATED-PARTICLE SEALED-TUBE NEUTRON PROBE: Q.S.T.I

DETECTION OF EXPLOSIVES, CONTRABAND, AND NUCLEAR MATERIALS

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# Associated-Particle Sealed-Tube Neutron Probe: Detection of Explosives, Contraband, and Nuclear Materials\*

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## *Abstract*

Continued research and development of the APSTNG shows the potential for practical field use of this technology for detection of explosives, contraband, and nuclear materials. The APSTNG (associated-particle sealed-tube generator) inspects the item to be examined using penetrating 14-MeV neutrons generated by the deuterium-tritium reaction inside a compact accelerator tube. An alpha detector built into the sealed tube detects the alpha-particle associated with each neutron emitted in a cone encompassing the volume to be inspected. Penetrating high-energy gamma-rays from the resulting neutron reactions identify specific nuclides inside the volume. Flight-times determined from the detection times of gamma-rays and alpha-particles separate the prompt (inelastic scatter and fission) and delayed (thermal-neutron capture) gamma-ray spectra and allow a coarse 3-D image to be obtained of nuclides identified in the prompt spectrum. The generator and detectors can be on the same side of the inspected object, on opposite sides, or with intermediate orientations. Thus, spaces behind walls and other confined regions can be inspected. Signals from container walls can be discriminated against using the flight-time technique. No collimators or shielding are required, the neutron generator is relatively small, and commercial-grade electronics are employed.

The use of 14- MeV neutrons yields a much higher cross-section for detecting nitrogen than that for systems based on thermal-neutron reactions alone, and the broad range of elements with significant 14-MeV neutron cross-sections extends explosives detection to other elements including low-nitrogen compounds, and allows detection of many other substances. Proof-of-concept experiments have been successfully performed for conventional explosives, chemical warfare agents, cocaine, and fissionable materials. Recent generator tests indicate that it should be practical to increase the source strength by up to an order of magnitude over the previous typical APSTNG operational levels, a benefit to time-critical applications. A small-footprint portal system with a gamma-ray detector array could be deployed for airport explosives and customs baggage and cargo inspection, or a complete single-detector system could be transported in an automotive van for on-site inspections.

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## 1. Present APSTNG System

A recently developed neutron diagnostic probe has potential for a range of van-mobile and fixed-portal applications for NDA (nondestructive analysis), including detection of explosives and drugs in aviation, customs, and physical security environments, arms control treaty verification, nonproliferation surveillance of SNM (special nuclear material) and CW (chemical warfare) agents, and characterization of radioactive waste and pollutants. The probe is based on an associated-particle sealed-tube neutron generator (APSTNG) that interrogates the object of interest with a low-intensity cone of 14-MeV neutrons generated from the deuterium-tritium reaction, detects the alpha-particle associated with each neutron, and records energy and flight-time spectra from gamma-rays resulting from neutron interactions in the object.

Investigations of applications for verification of chemical and nuclear weapons [1], detection of explosives and drugs [2,3], and extension to large interrogation volumes [3,4] are covered in previous publications. Most recently, application studies have been conducted for detection of cocaine in propane tanks, monitoring for smuggled plutonium and uranium, and characterization of radioactive and toxic waste. A new advanced APSTNG (along with improved high voltage supply and control unit) has been designed and is being constructed. This paper is a review of ANL investigations of this technology.

### 1.1 Principles of operation

The neutron diagnostic probe which Argonne has been using was developed primarily by Nuclear Diagnostic Systems (NDS) [5]. Its operation can be understood from Fig. 1, which sketches a "general purpose" APSTNG system. The object to be interrogated might be baggage or cargo or an item to be inspected under arms control treaty provisions or radioactive waste regulations, that could contain explosives, illicit drugs, chemical warfare or other toxic agents, or fissionable or radioactive material. In the APSTNG, deuterons are accelerated into a tritium target, producing 14-MeV neutrons isotropically. Each neutron is accompanied by an associated alpha-particle travelling in the opposite direction (in center-of-mass coordinates). The gamma-ray and neutron detectors are time-gated by

pulses from the alpha detector, forming a cone of flight-time-correlated neutrons through the object. Detector pulses are time-resolved by CFD's (constant-fraction discriminators). Flight times are determined by a TAC (time-to-amplitude converter), digitized by an ADC (analog-to-digital converter), and recorded.

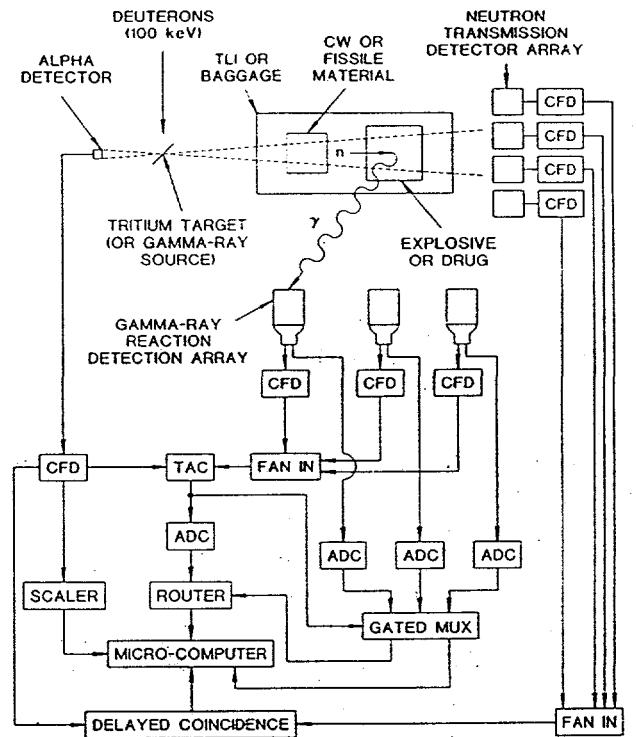


Fig. 1. Schematic layout of "general purpose" APSTNG-based interrogation system, with EGRIS and FNTI modes.

A single specially configured gamma-ray detector is employed in the current ANL APSTNG system. A NaI(Tl) crystal 10.16 cm  $\times$  10.16 cm in cross section and 40.64 cm long is coupled to two fast photomultipliers, one at each end. Improvements in time and energy resolution are obtained by combining the timing and energy data from the two ends, while the large detector provides relatively high efficiency for the high-energy gamma rays.

When a reaction occurs in the object along the cone that results in a detected gamma-ray, the time-delay from the alpha pulse yields the position (depth) along the cone where the reaction occurred, since the source neutron and gamma-ray speeds are known (5 cm/ns and 30 cm/ns, respectively). By using a two-

dimensional (2D) position-sensitive multipixel alpha detector, transverse and depth coordinates of reaction sites can be mapped, providing three-dimensional (3D) emission imaging of reaction densities from measurements at a single orientation. (The present NDS system uses a PM tube that is not position-sensitive to detect alpha scintillations, but can provide 3D imaging in the laboratory by restricting the alpha window view field and scanning the interrogated object transversely.) Depth resolution along the cone axis is limited by the  $\sim 1$  ns time resolution of the flight-time electronics and detector signal pick-off to no better than  $\sim 5$  cm.

In applications requiring imaging, systems would include a 2D alpha detector, as well as an array of gamma-ray reaction detectors, so as to maximize information obtained from each interacting neutron and sufficiently minimize measurement time. The PC controls the experiment, calculates positions, and displays data and images. Software can be developed for specific applications that will perform intelligent data analysis and interact with the operator to determine which items are sufficiently suspect to require further examination.

## 1.2 Detection modes

Fast-neutron inelastic scattering reactions in the object provide prompt gamma-ray spectra that can identify many nuclides. By choosing gamma lines of specific nuclides, a coarse 3D image of each identifiable nuclide in the time-correlated spectrum can be mapped. By choosing appropriate nuclide intensity ratios, 3D images of compounds can be made (molecular bonds are not identified). The use of the time-correlated gamma-ray spectra is denoted the EGRIS (emissive gamma-ray imaging and spectroscopy) mode. If fissionable materials are present, neutron detectors may be used to detect emitted fission neutrons in the ENIS (emissive neutron imaging and spectroscopy) mode. Nearly all nuclides with atomic number above boron have distinctive gamma-ray spectra for the EGRIS mode, with reaction cross sections of  $\sim 0.5$  barn for 14-MeV neutrons (predominantly inelastic scattering).

For gamma-rays above  $\sim 1$  MeV, background is greatly reduced since background counts can only be accumulated during the nanosecond-range correlation interval.

Because neutrons are emitted isotropically, the source and emission detection systems can be located arbitrarily, and can be on the same side of the interrogated object when access is restricted. Regions behind walls, under floors or roadbeds, or above ceilings could be inspected nonintrusively. The high-energy neutrons and gamma-rays will penetrate large objects and dense materials. The EGRIS mode is generally the primary detection mode.

Slow-neutron capture is not time-correlated with the alpha pulses, but provides nonimaging gamma-ray spectra that can aid nuclide identification. Use of non-correlated gamma-ray spectra with the neutron generator on is termed the CGRS (capture gamma-ray spectroscopy) mode. CGRS data can be collected simultaneously with EGRIS data by using a multi-channel analyzer board inside the PC. The thermal neutron capture cross section is small for most nuclides, but is large for nuclides of interest in some applications. The gamma-ray spectra are generally substantially more complex than for EGRIS mode, with much more background. Neutron moderator material may need to be placed between the APSTNG and the interrogated object to get sufficient intensity.

Another detection mode of use is the PGRS (passive gamma-ray spectroscopy) mode, for which the neutron generator is turned off, allowing detection of gamma-ray radioactivity present in the interrogated object and in its vicinity, including any activity induced in the object by the neutrons as well as uncorrelated background. This mode is used for static system energy calibration with gamma-ray sources (dynamic energy calibration is performed in the EGRIS mode using known materials and their reaction gamma-ray spectra). Because cross sections for inducing activity with a finite half-life (several seconds or more) are usually very small, the PGRS mode is generally used only when gamma-ray radioactivity is known to be present (as for radioactive waste or nuclear weapons).

As shown in Fig. 1, by discarding detected neutrons not having the proper flight time to be uncollided, one can perform fast-neutron 2D transmission imaging without a collimator (by scanning, using a neutron detector hodoscope array, or using 2D neutron detectors), since scattered neutrons are removed by "electronic collimation". This is called the FNTI (fast-

neutron transmission imaging) mode. By measuring at a sufficient number of views around 180 degrees, 3D tomography with relatively coarse spatial resolution is feasible. Transmission imaging (FNTI) can be done along with or instead of emissive reaction-density imaging (EGRIS). No spectral distinction between nuclides is provided, but the neutron attenuation coefficient is mapped over the interrogated object. The FNTI mode is currently used to map and position the neutron correlation cone, and may find use for neutron attenuation corrections or for inspecting extended or highly absorbing objects [3,4].

### 1.3 The NDS sealed-tube neutron generator

As diagrammed in Fig. 2, a Penning ion source inside the NDS APSTNG emits a continuous mixed beam of deuterium and tritium ions that is accelerated and focused on a small spot (~ 1 mm diameter) on the target, tritiatating the target and producing neutrons and alpha particles. (The small spot is necessary to obtain good spatial resolution.) A zirconium getter controls the mixture of deuterium and tritium. The ions are accelerated by a high voltage of 95 Kv and focused by a variable extraction voltage, nominally 15 Kv. The ion beam current (~ 1  $\mu$ A to get  $10^6$  n/s) is controlled by varying the getter heating current. All operating voltages and currents are provided and monitored by a high-voltage control system. The welded metal-ceramic tube contains 0.4 Ci tritium at low pressure compared to the atmosphere, and the tritium is contained in the getter when the tube is not operating. The single-pixel alpha detector consists of a ZnS screen inside the tube, with a photomultiplier outside interfaced to a window. (For a multipixel 2D alpha detector, the photomultiplier can be replaced by a microchannel plate and matrix anode readout, as shown in Fig. 2.) The alpha scintillator is limited to materials that can withstand tube bake-out temperatures during manufacture. ZnS has high light output and is inexpensive.

Initial maximum output of a typical NDS APSTNG is around  $3 \times 10^7$  n/s, but the maximum output soon falls to about the level of  $10^7$  n/s, as the cathode target is sputtered away, and slowly decreases thereafter. An output of ~  $10^6$  n/s can be maintained for ~ 2000 hours by increasing the ion current to compensate for sputtering. Usable neutron source strength can be

limited by accidental gamma-ray counts, which increase as the square of the count rate and become larger in number for large interrogation volumes.

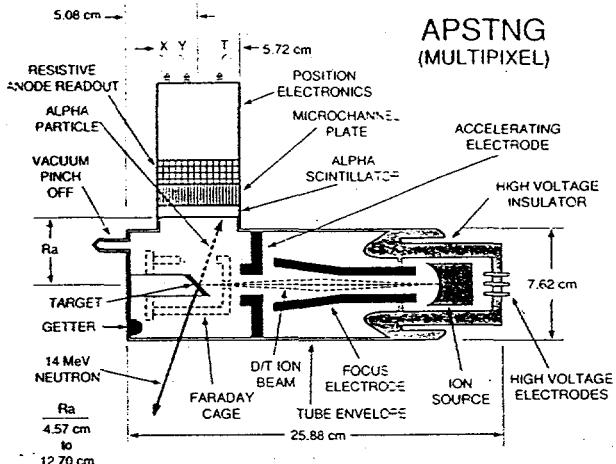


Fig. 2. Cross-section of NDS-type APSTNG tube, with 2D position-sensitive multipixel alpha-particle detector.

The design of the APSTNG differs substantially from the current well-logging neutron generator tubes, which cannot be used for associated-particle operation. (Well-logging tubes are usually pulsed, have no capability to focus the ion beam on a small spot on the target, and have no internal scintillator.) The APSTNG is a relatively inexpensive small sealed module with low-bulk support equipment. It is easily replaced, presents low radiation exposure, and the sealed-tube design prevents tritium contamination.

## 2. Recent Investigations of Potential Applications

Most recently, specific APSTNG applications have been studied for detection of cocaine in propane tanks, monitoring for smuggled plutonium and uranium, and characterization of radioactive and toxic waste. Proof-of-concept laboratory experiments using the present NDS APSTNG system, some benchmark experiments, and sensitivity calculations for fielded prototype APSTNG systems based on experiment data have been performed. Unless otherwise noted, in the experiments the gamma-ray detector is far enough from the interrogated object to separate scattered neutron background by flight time and the alpha-particle window aperture is set to fully cover the interrogated objects.

## 2.1 Detection of cocaine in propane tanks

The application is the detection of cocaine hidden inside large liquid propane tanks, falsely declared by the shipper to be either "empty" or "full of propane". Detection of cocaine inside a tank declared to be empty is straightforward, because a tank that is really empty would exhibit only the signature of the tank wall composition (observation of any other gamma rays would arouse suspicion). Discriminating between cocaine and propane is more challenging, so a direct demonstration of discrimination between cocaine and propane was performed.

Cocaine hydrochloride ( $C_{17}H_{22}Cl N O_4$ ) is the form most likely to be smuggled, rather than free-base cocaine ( $C_{17}H_{21} N O_4$ ). The elemental composition of propane is  $C_3H_8$ . Thus, cocaine detection may be focussed on detecting the presence of one or more of the elements chlorine, nitrogen, and oxygen. Chlorine has major gamma-ray peaks at 1.22, 1.76, and 2.15 MeV, while nitrogen has major peaks at 1.63, 2.31, 4.46, 5.10, and 7.03 MeV. Oxygen has gamma rays from 2.74 to 7.12 MeV, with a major peak at 6.13 MeV (giving rise to single-escape and double-escape pair production peaks at 5.62 and 5.11 MeV). Hydrogen has no inelastic scattering. The carbon signature is the 4.43 MeV gamma-ray peak plus single-escape and double-escape pair production peaks at 3.92 and 3.41 MeV. The signature of iron in the steel container is gamma-ray peaks at 0.85 and 1.24 MeV.

Polyethylene ( $C_2H_4$ ) was selected as the liquid propane simulant, as beads of density  $0.576 \text{ g/cm}^3$ , yielding a carbon density of  $0.492 \text{ g/cm}^3$ . This compares favorably with propane, with density  $0.501 \text{ g/cm}^3$  and carbon density of  $0.410 \text{ g/cm}^3$ . "Elemental" surrogates for cocaine and cocaine hydrochloride can be made from commercially available plastic feedstocks and carbon. To duplicate the relative elemental densities of cocaine HCl, we chose to mix one mole each of polyacrylonitrile, polyvinyl chloride, and polymethyl methacrylate with 1/2 mole each of poly(1,4 butylene terephthalate) and polyethylene.

The materials were separately ground to a small grain size in a blender with dry ice. The polyethylene was tough and required further milling to get a relatively small grain size. The materials were then mixed. In

order to ascertain the actual composition and composition variation of the surrogate, chemical analyses of samples from the top, middle, and bottom of the container were performed. The results indicate the surrogate mix is uniformly distributed and is an excellent "elemental" simulant of cocaine HCl.

The cocaine HCl surrogate in its cylindrical bottle has a net density of  $0.709 \text{ g/cm}^3$ , with a carbon density of 0.426 (similar to propane, so the carbon signals will approximately cancel), an oxygen density of 0.133, a chlorine density of 0.074, and a nitrogen density of 0.029. Thus, we would expect the oxygen to be easiest to detect, the chlorine to be harder to detect, and the nitrogen to be hardest to detect (everything else being equal). Measurements were made with small samples compared to those expected in the actual application, 718 g for the cocaine hydrochloride simulant and 671 g for the liquid propane simulant. Each sample was enclosed in a steel cylinder 30.5 cm high, 20.3 cm o.d., with 0.64 cm wall to simulate the tank. The neutron source rate was  $\sim 4 \times 10^6 \text{ n/s}$ .

Figure 3 shows the result of subtracting the normalized energy spectrum of simulated propane from that of simulated cocaine hydrochloride. (This subtraction results in approximately zero net counts in the carbon peaks, providing a check that the normalization was carried out properly.) The net spectrum clearly shows the presence of chlorine, nitrogen and oxygen in the cocaine hydrochloride simulant. The high-energy oxygen peaks are prominent.

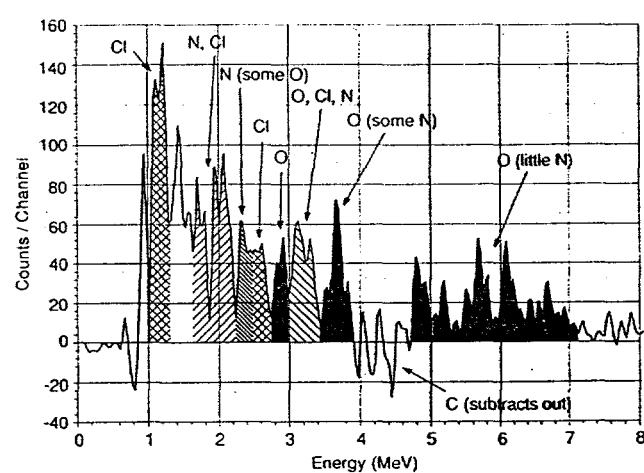


Fig. 3. Detection of cocaine hydrochloride in propane by EGRIS spectrum subtraction.

Counting time for each of these two measurements on relatively small samples was 60 minutes. Analysis of the data indicates that 0.7 kg of cocaine HCl can be distinguished from propane with  $1-\sigma$  statistics in only 2 minutes with our single-detector system. In previous studies of drug detection in luggage and cargo containers [3,4], the relatively small amount of oxygen (in particular the low O/C ratio) was a distinguishing signature for drugs, whereas in this case oxygen provides the most prominent signature (since oxygen is easily detected by the APSTNG and is not present in propane).

## 2.2 Monitoring for smuggled plutonium and uranium

14-MeV neutrons stimulate emission of both prompt fission neutrons and prompt fission gamma rays from fissionable materials, so both the EGRIS and ENIS modes could be employed for identification. Because the spectra are nearly identical in shape for all the isotopes of uranium and plutonium, these modes cannot, however, by themselves distinguish between isotopes (but this distinction may not be necessary, depending on scenario). Actinide inelastic scattering gamma rays are too low energy to be detected reliably against background in the EGRIS mode. The individual isotopes could be distinguished by the CGRS mode, but thermal neutrons would penetrate actinide materials very poorly.

In the 1-3 MeV detectable range,  $\sim 2.4$  gamma rays are emitted per fission. The U-238 cross section for fission by 14-MeV neutrons is  $\sim 1$  barn, about half that for U-235. Figure 4 shows the EGRIS prompt gamma-ray spectrum from APSTNG-induced fission in U-238. EGRIS spectra measured for enriched U are similar to Fig. 4, but without the two superimposed peaks. These two small peaks result from the U-238 daughter Pa-234m decay lines at 0.766 and 1.001 MeV manifested as background accidental coincidences because of the substantial amount of U-238 present, yielding a (nonimaging) signature for U-238.

We consider a generic application in which a checkpoint inspection system is set up to monitor for undeclared fissile material. The APSTNG system is configured to inspect items simultaneously for unshielded U-235 and Pu-238 in a passive mode, and for the presence of shielding material in the EGRIS

mode. An item that gives a positive response to the passive radiation inspection would be set aside for additional inspection, including high-resolution x-radiography and APSTNG measurements to detect other materials. An item that gives a positive response for presence of shielding materials, but no passive measurement of gamma-ray signatures for U-235 or Pu-239, would be x-rayed and would be counted by the APSTNG for an extended period of time to compensate for extensive shielding.

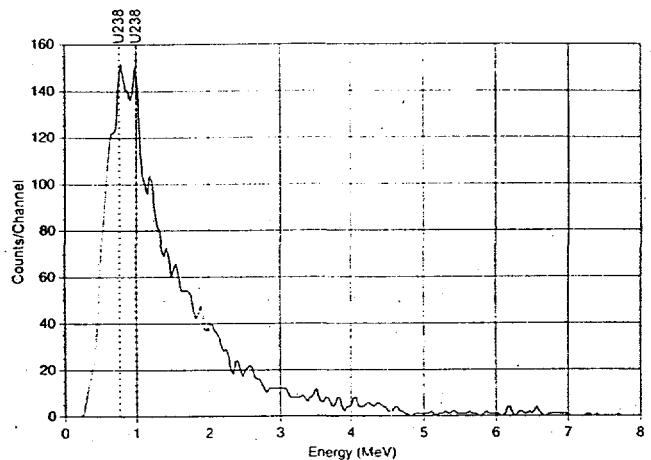


Fig. 4. EGRIS energy spectrum measured for U-238 fission.

The APSTNG system proposed for this application would employ an inspection volume with a radiation detector array on each of three sides. The reference inspection object is a closed cube 30 cm on a side. Each array would consist of 4 gamma-ray detectors, each of the present double-ended NaI type, with a total array active area of  $40 \text{ cm} \times 40 \text{ cm}$ . In order to avoid interference from neutron scattering, each array would be positioned 70 cm from the center of the interrogated object, and the neutron generator, running at  $\sim 10^8 \text{ n/s}$ , would be placed 50 cm from the object center.

In a lightly shielded case (item surrounded by 1 cm Pb, sufficient to defeat passive detection of the 186-keV U-235 gamma), 750 g of Pu or U should be detected in the EGRIS mode within 30 seconds, or 15 g within 15 minutes. In a heavily shielded case (item surrounded by 5 cm Pb, sufficient to defeat passive gamma-ray detection of Pu-239 and U-235, plus 6 cm borated plastic, sufficient to cloak the material from active detection by a fission neutron source such as Cf-252,

but insufficient to exclude 14-MeV neutrons), 750 g of Pu or U should be detected within 30 minutes. Conventional high explosives would be detected by the proposed APSTNG system in less time than required for the Pu and U.

### 2.3 Characterization of radioactive and toxic waste

The APSTNG capability to coarsely determine locations and shapes of fissionable materials and toxic chemicals by EGRIS measurements of fission and inelastic gamma rays could be useful in characterizing radioactive waste in assay of soils and drums. Current radwaste assay methods focus on high-energy x-radiography, active neutron interrogation for detection of fissile material, and passive measurements of radiation, rather than material-specific location or identification of non-radioactive elements or chemicals. An APSTNG system could supplement other technologies in characterization of waste containers. If attenuation by materials in the container is small, passive radiation detection will be substantially more sensitive than active neutron interrogation. If attenuation is large, the opposite will be true; for example, the 186-kev U-235 gamma ray and the 414-kev Pu-239 gamma ray are much easier to attenuate than the prompt fission gamma rays from APSTNG interrogation. The use of active neutron interrogation for identification of toxic chemicals, such as chlorinated compounds, can help classify the radwaste as to mixed waste content, and the ability to detect local regions of water (by detection of oxygen) may address criticality issues.

ANL has fabricated a radwaste calibration drum based on calibration drums from the Stored Waste Examination Pilot Plant (SWEPP), at the Idaho National Engineering Laboratory (INEL) Radioactive Waste Management Center (RWMC). It is a 55-gallon steel drum of diameter 58 cm and length 88 cm, with three aluminum tubes of 84-mm i. d. positioned at different radii running the length of the drum, to allow samples to be inserted inside the drum. The top third of this drum is a projection of the cross-section of the SWEPP heterogeneous mixed metals calibration drum. The central third of the ANL drum is left void, to provide space for future configurations. The bottom third of the drum is filled with concrete mortar, similar to a SWEPP concrete sludge calibration drum.

A LiCl sample of ~ 400 g in a 102 mm high bottle of 76 mm diameter and a depleted U sample 25.4 mm square and 127 mm high were placed inside tubes of the ANL radwaste calibration drum that were aligned upright such that a cross section of each tube intercepted the neutron correlation cone axis. The APSTNG neutron rate was kept near  $6 \times 10^6$  n/s and the runs were 1 hour long.

Data from a representative measurement in the bottom concrete sludge section is shown in Fig. 5, the EGRIS energy spectrum for flight-time channels 60-93, corresponding to the position of the LiCl sample in the tube nearest the drum edge. Evident are the chlorine gamma-ray peaks from the sample and oxygen gamma-ray peaks from  $\text{SiO}_2$  (sand) and residual water in the surrounding concrete mortar. Shown as a dashed line is a residual fission gamma spectrum from the depleted U, nearby in the center tube. The EGRIS energy spectrum for flight-time channels 93-119 (not shown), corresponding to the position of the depleted U sample, has a dominant fission gamma-ray spectrum.

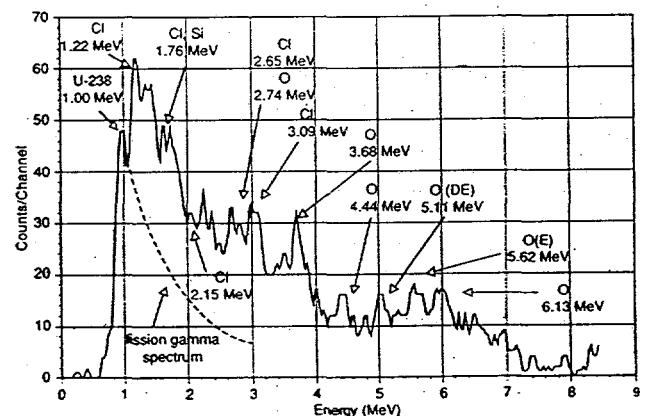


Fig. 5. Chlorine and depleted uranium in concrete sludge section of ANL radwaste calibration drum. EGRIS energy spectrum for flight-time channels 60-93.

Shown in Fig. 6 is the PGRS spectrum obtained for a SWEPP radwaste sludge drum, where the signal run is designated in black and the background run is designated in gray (the background run was only 100 seconds long). The Pu-239 peak group is quite evident (individual peaks are not resolved due to the limited energy resolution of NaI). Background peaks are seen from Am-241, K-40, and U-232 or Th-232 (the 2614 keV gamma ray comes primarily from Tl-208,

which can be a daughter of either U-232 or Th-232, but here the parent isotope almost certainly is Th-232.) The PGRS spectrum for a drum found to contain U-238 looks similar, except that the Pu-239 peak group is replaced by the 766 and 1001 keV peaks from the Pa-234m daughter of U-238.

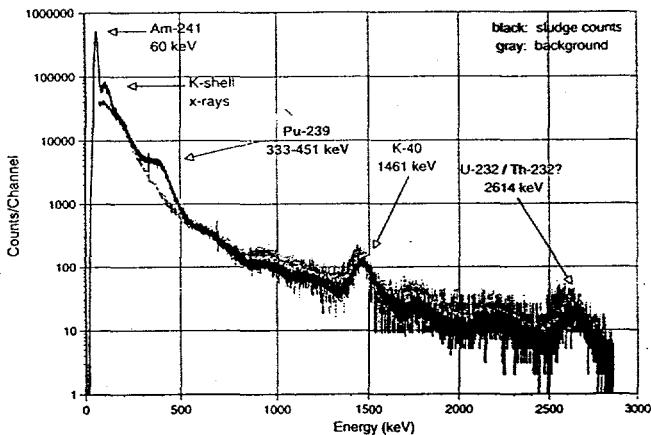


Fig. 6. Passive gamma-ray spectrum of RWMC radwaste sludge drum RF074404275.

#### 2.4 Identification of chemical warfare agents

Early investigations of APSTNG detection capability for CW agents and munitions were promising, including an analytic study of gamma-ray energies and cross-sections for neutron inelastic scattering and element structure of CW agents and explosives, as well as measurements taken of 155-mm shells filled with CW simulants using an early version of the APSTNG system [1,2]. Recently we have measured spectra of the basic elements involved as well as of new CW surrogates we have prepared for some nerve and mustard agents, using our large double-ended NaI detector. Our results indicate that CW munitions can be distinguished from conventional high-explosive munitions, but that an HPGe detector, with its high energy resolution, may be needed to supplement our NaI detector for distinguishing between CW agents having overlapping closely spaced lines in their gamma-ray spectra.

As illustrated in Fig. 7, simultaneous identification and imaging of individual munitions in cartridge boxes and pallets, without requiring opening and breaking out of the rounds for separate measurements, is a potential EGRIS-mode advantage in CW detection, as us

sensitivity to partial fills of CW munitions. The coarse APSTNG imaging capability could lead to a substantial throughput increase in CW munition verification and to more accurate monitoring for CW demilitarization operations.

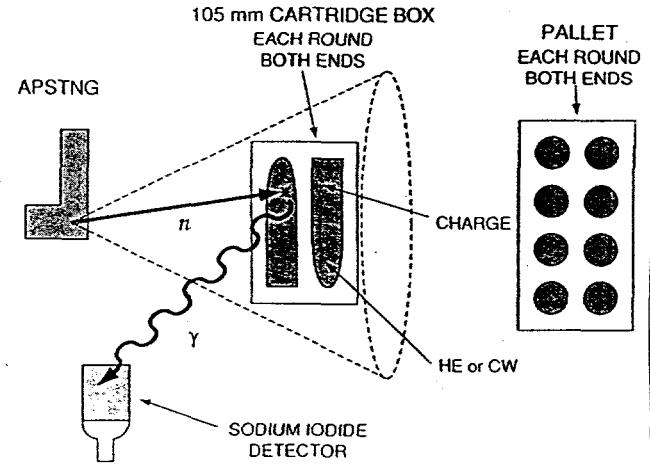


Fig. 7. Identification and imaging of CW munitions.

#### 3. Portal Monitor for Explosives and Drugs

Based on data from APSTNG laboratory experiments on explosives and drugs, it is interesting to estimate the measurement times required for detection of contraband items in some simple idealized cases using a conceptual APSTNG portal monitor system (such as might be deployed for airport luggage inspection), in this case, one having a multipixel alpha-particle detector and eighteen relatively large double-ended NaI gamma-ray detectors, as shown in Fig. 8. The laboratory data consists of EGRIS gamma-ray spectra for nitrogen, oxygen, and carbon, for specific explosives (C-4, PBX, Octol, Composition B, TNT, Pentolite, Dynamite, and Deta sheet), and for cocaine and heroin simulants.

The spectrum for a typical explosive, such as C-4 shown in Fig. 9, exhibits a number of principal peaks (mostly from neutron inelastic scattering) characteristic of nitrogen, oxygen, and carbon, along with a number of escape peaks, with a number of peaks overlapping. (In Fig. 9, "SE" and "DE" refer to single and double escape peaks, respectively.) The proportion of nitrogen is relatively high for conventional explosives, compared to other materials likely to be present (the density of explosives tends to be high also). The

spectrum for a typical narcotic, such as cocaine (shown in Fig. 10) and heroin [4], tends to have a high proportion of carbon compared to oxygen.

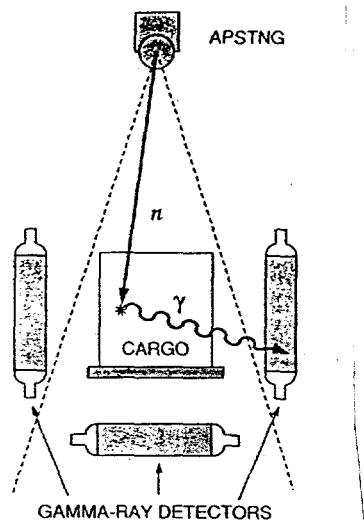


Fig. 8. Design concept of EGRIS portal monitor, end view. Five more detectors are behind each double-ended gamma-ray detector shown (PM tube at each end), a total of 18 detectors, each 15 cm  $\times$  15 cm  $\times$  45 cm.

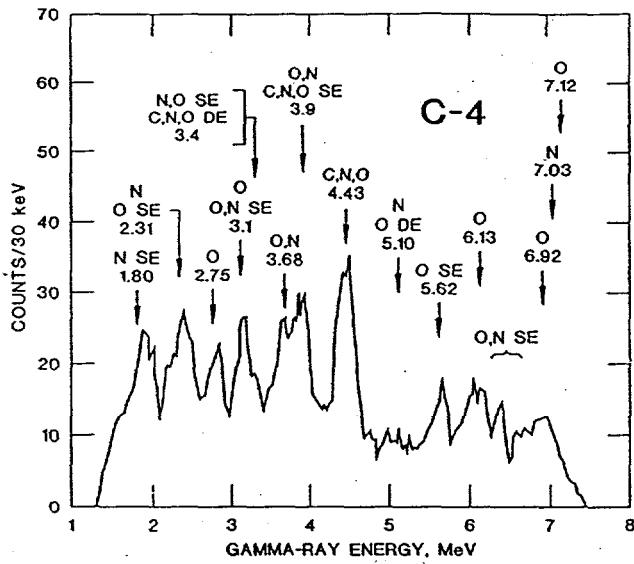


Fig. 9. EGRIS gamma-ray spectrum of C-4 explosive.

By a judicious choice of energy windows and knowledge of the basic nitrogen, oxygen, and carbon spectra, a "detection" algorithm was obtained that correctly reproduces the density ratios of nitrogen to oxygen and carbon to oxygen for the explosives and for other materials that might be present. It was found that a plot of these ratios is useful for differentiating

contraband drugs or explosives from ordinary items expected to be seen (see Fig. 11). Materials that would normally appear in luggage or foodstuffs are separated from explosives (open squares) and cocaine.

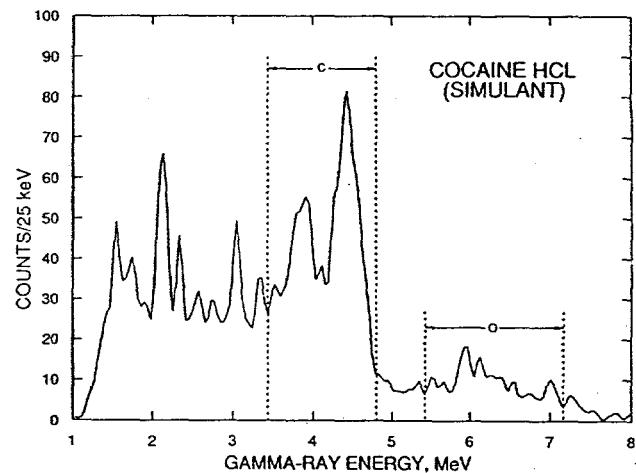


Fig. 10. EGRIS spectrum of cocaine HCl simulant.

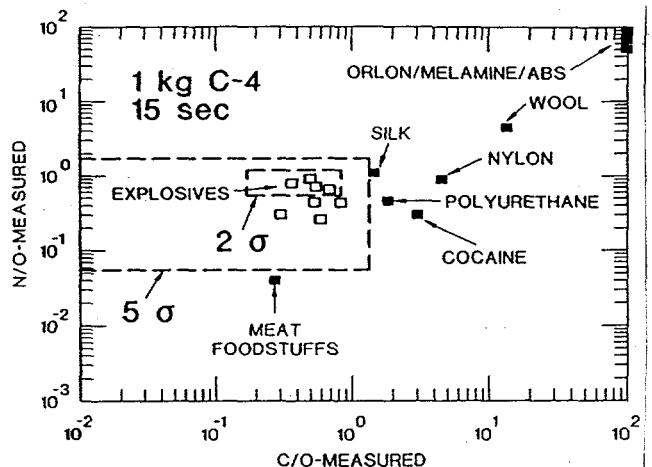


Fig. 11. N/O vs. C/O for 15-s measurement of 1 kg C-4. Large box envelops 5- $\sigma$  statistics; small box envelops 2- $\sigma$ .

The boxes shown in Fig. 11 represent count statistics for a 15-second measurement of 1 kg C-4 explosive, the larger box enveloping five standard deviations (5 sigma), the smaller box enveloping two standard deviations. (The C-4 is assumed to be located in one volume element, rather than being spread out in a sheet.) The C-4 is definitely identified as an explosive. Even a 4-second measurement would distinguish C-4 from items normally found in luggage, so if luggage is being examined, suspicion is indicated. In 480 seconds of measurement time, the C-4 would be differentiated from other common explosives.

In Fig. 12, boxes are drawn for two and five standard deviations of count statistics for a 2-second measurement of 1 kg of meat. With high probability, the meat is identified as a foodstuff rather than explosives or contraband, indicating a very low false alarm rate for monitoring foodstuffs. In Fig. 13, a two-standard-deviation box is shown for a 4-second measurement of 1 kg of cocaine in fish. (The cocaine is assumed to occupy only one volume element.) The item is identified as suspicious, since it is not just fish, and is thought probably to be cocaine, since it is not amphetamines (it could be plastic, but plastic in fish would be a bizarre, rare, suspicious event).

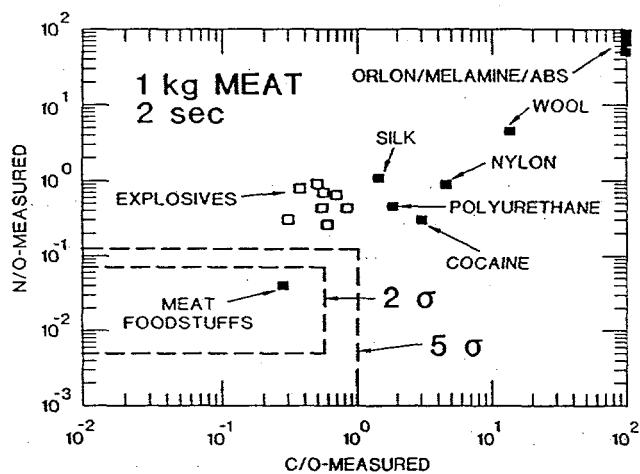


Fig. 12. N/O vs. C/O for 2-s measurement of 1 kg meat. Large box envelops 5- $\sigma$  statistics; small box envelops 2- $\sigma$ .

These simple computations and the underlying laboratory data can be considered as comprising a feasibility study for the application of an APSTNG system to monitoring contraband explosives and drugs, but are of course no substitute for analysis of laboratory measurement of realistic mockups and actual field trials of prototype units. The presence of noncontraband materials (attenuation and scattering effects and the complex arrangements of many diverse items that may normally appear) and deception attempts (such as explosives and drugs in sheet form, shielding, judicious placement of noncontraband materials, and adulteration of drugs) have not been considered. On the other hand, much more powerful detection algorithms can be developed, that utilize material densities directly along with full spectral regression fitting, adaptive learning processes, and external information from other surveillance methods.

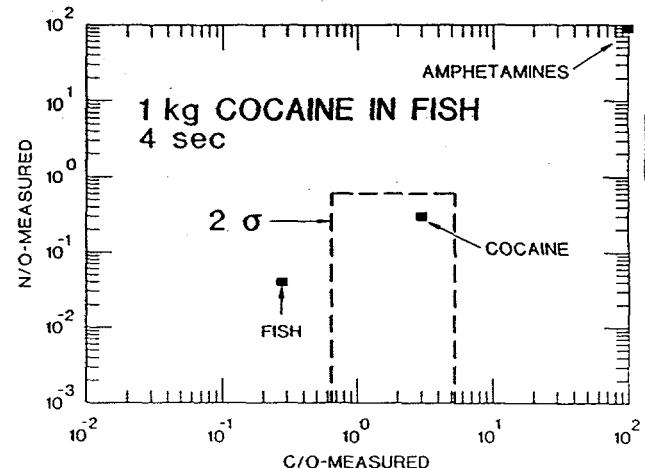


Fig. 13. N/O vs. C/O for 4-s measurement of fish containing 1 kg of cocaine. Box envelops 2- $\sigma$  statistics.

APSTNG portal monitoring systems for cargo container and truck inspection are beyond the scope of this paper, but are described in Ref. 4. Multiple APSTNG systems are required for large inspection volumes. A two-stage inspection arrangement in which the first stage searches for "suspicious" cargo over the full container and a second stage identifies contraband rather precisely for offloaded suspicious cargo should be cost-effective in recovering most of the contraband and in reducing labor in offloading-reloading and unpacking-packing caused by false alarms.

#### 4. Advanced APSTNG System

In order to meet field criteria for a number of important applications, it is necessary to develop an APSTNG system of more advanced design. The NDS-type APSTNG system has proved itself in the laboratory, but additional improvements will be needed. Although the NDS neutron tube proved to be reliable, time critical applications demand higher neutron output and longer life in terms of integrated neutron output, and field use requires more rugged construction, particularly a rugged accelerator head and HV coupling. The NDS HV control unit initially performed satisfactorily, but as it aged, it began to malfunction, giving spurious meter readings and experiencing repetitive HV breakdown and arcing, apparently creating voltage and current surges in the neutron tubes that caused them to fail.

ANL is collaborating with MF Physics on a new higher-output longer-life sealed-tube neutron generator and an improved control unit with HV supply, designed to be rugged and transportable and making good use of lessons learned with the NDS APSTNG system. These components will interface directly with the existing ANL single-pixel single-detector system.

MF Physics is designing and building the basic sealed tube to be a welded metal-ceramic unit that can withstand mechanical vibrations and shocks during van transportation. A drawing of this new APSTNG tube is shown in Fig. 14, where cutaways reveal the alpha window and beam forming lens configurations. Ceramic parts have been subjected to drop tests to assure ruggedness. The tube is being outfitted with two getters of an advanced design, with one acting as a backup for the other (the present NDS tube has one getter of simple design that can be damaged by overheating).

MF PHYSICS APSTNG

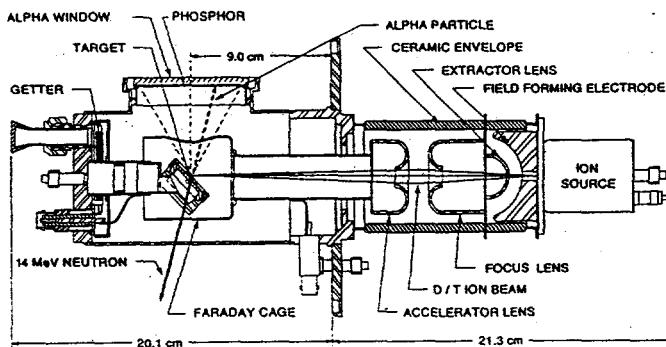


Fig. 14. Cut-away design drawing of new advanced APSTNG constructed by MF Physics.

MF Physics will warranty the unit to provide a neutron output of at least  $10^8$  n/s without any target or ion source cooling, for a summed total operating time of at least 800 hours, and a maximum output of at least  $10^9$  n/s with externally supplied target water cooling (circulation of an externally supplied freon-like coolant around the ion source may also be required for  $10^9$  n/s). The maximum continuous output rate with no cooling is  $\sim 10$  times that for the NDS APSTNG tube, and the number of neutrons generated during the expected lifetime is  $\sim 40$  times that expected for an NDS APSTNG tube. More than an 8000-hour lifetime

at the  $10^7$  n/s output rate needed for many applications is expected and the warranted neutron generation is sufficient for field use in any application considered. (Also, a spare tube can be kept on hand for immediate replacement, if desired.)

For the planned alpha window solid angle, alpha count rates could reach  $\sim 7 \times 10^6$  per second at  $10^8$  n/s and  $\sim 7 \times 10^7$  per second at  $10^9$  n/s, so that the alpha window scintillator should have an effective mean light decay on the order of 50 ns or less for  $10^8$  n/s or 5 ns or less for  $10^9$  n/s, with no significant long-persistence light tails, in order that pulse pileup and saturation effects are minimal. The alpha scintillator rise time should be in the subnanosecond range, in order to maximize flight-time resolution. Unfortunately the ZnS(Ag) alpha scintillator used in the present NDS neutron tubes has a long 200-ns mean decay time.

Other important alpha scintillator properties include luminosity, transmission of emitted light, resistance to radiation damage by neutrons and alpha particles, ability to withstand bake-out at up to 400 C in a reducing atmosphere, availability as large-diameter thin crystals or in a grain-size range allowing relatively uniform deposition of appropriate thickness, and emission wave-band (wavelengths shorter than  $\sim 365$  nm will not be transmitted by fiber-optic windows). Many scintillators have been investigated, and ZnO(Ga) has been chosen for use. Ga doped ZnO is the fastest scintillator available, having a decay time of 0.7 ns, and it provides moderate luminosity in the 365-450 nm range. ZnO(Ga) meets most of the requirements well, but absorbs its emitted light, so coating thickness and uniformity may be critical.

The present NDS HV coupling unit is based on large load-bearing O-ring seals (that tend to leak under relatively light stress) and a housing containing Fluorinert insulating fluid that must be drained before the neutron tube can be transported. The HV coupling unit for the new neutron tube (see Fig. 15) will mate to a flange welded onto the accelerator tube with large machine screws, such that the coupling housing is the load bearing surface in a rigid mount. O-rings bear no structural loads and are used only for sealing in pressurized SF<sub>6</sub> insulating gas. The unit will have a pressure gage and a pressure switch that provides a low-pressure interlock and panel warning light.

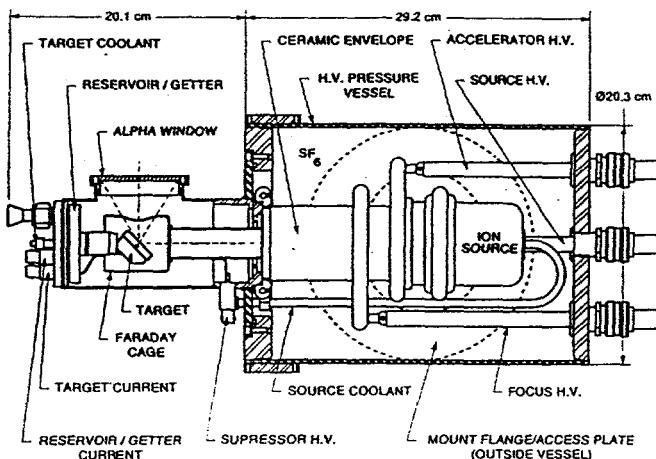


Fig. 15. Design drawing of accelerator head for advanced APSTNG constructed by MF Physics.

The new HV supply and accelerator control system will protect the APSTNG neutron tube against voltage and current surges with limiting circuitry, and a remote HV-kill switch will be provided that can be carried by an operator for shutdown in case of any emergency or malfunction in the system. The design of the new control unit and state-of-the-art HV supplies, with a separate supply for each voltage terminal, provides a simpler control system than the NDS unit that is much less likely to malfunction and unlikely to be susceptible to repetitive arcing as the system ages.

A relatively large HPGe gamma-ray detector has been procured, in order to provide the high energy resolution needed in applications defined above that involve complex gamma spectra having overlapping peaks in data from the NaI detectors. This detector has been specially configured to maintain high energy resolution during neutron irradiation and to minimize down-time due to radiation damage. It will work with the present APSTNG data acquisition system (and the new neutron generator tube), with some limitations. Future developments for specific applications might include an array of multiple detectors, multipixel alpha detector, and a new data acquisition system.

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