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APSTNG: Neutron Interrogation for Detection of Nuclear and CW Weapons, Explosives, and Drugs

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ABSTRACT

A recently developed neutron diagnostic probe system has the potential to satisfy a significant number of van-mobile and fixed-portal requirements for nondestructive verification of sealed munitions and detection of contraband explosives and drugs. The probe is based on a unique associated-particle sealed-tube neutron generator (APSTNG) that interrogates the object of interest with a low-intensity beam of 14-MeV neutrons generated from the deuterium-tritium reaction and that detects the alpha-particle associated with each neutron. Gamma-ray spectra of resulting neutron inelastic scattering and fission reactions identify nuclides associated with all major chemicals in chemical warfare agents, explosives, and drugs, as well as many pollutants and fissile and fertile special nuclear material. Flight times determined from detection times of the gamma-rays and alpha-particles yield a separate tomographic image of each identified nuclide. The APSTNG also forms the basis for a compact fast-neutron transmission imaging system that can be used along with or instead of the emission imaging system; a collimator is not required since scattered neutrons are removed by "electronic collimation" (detected neutrons not having the proper flight time to be uncollided are discarded). The small and relatively inexpensive APSTNG exhibits high reliability and can be quickly replaced. Proof-of-concept experiments have been performed under laboratory conditions for simulated nuclear and chemical warfare munitions and for explosives and drugs.

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1. Introduction

New developments in hodoscope radiation detection and neutron diagnostic probe technologies offer some rather unique capabilities for a wide range of applications, including arms control treaty verification, remediation of radwaste and pollutants, and detection of explosives, drugs, and other contraband. In the hodoscope concept, an array of radiation detectors is used to image or detect objects inside opaque containers. Argonne interest stems from the fast-neutron hodoscope [1] in use at the Transient Reactor Test (TREAT) Facility for many years to image motion inside thick steel capsules of reactor fuel undergoing destructive testing that simulates hypothetical core-disruptive accidents.

Gamma-ray and neutron hodoscopes can be combined with a recently developed neutron diagnostic probe to satisfy a wide range of van-mobile and fixed-portal applications for NDA (nondestructive analysis). The probe is based on a unique associated-particle sealed-tube neutron generator (APSTNG) that interrogates the object of interest with a low-intensity beam of 14-MeV

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neutrons generated from the deuterium-tritium reaction and detects the alpha-particle associated with each neutron. Gamma-ray spectra of resulting neutron reactions identify nuclides associated with all major chemicals in CW (chemical warfare) agents, explosives, and drugs, as well as many pollutants and fissile and fertile SNM (special nuclear material). Flight times determined from detection times of the gamma-rays and alpha-particles yield a separate low-resolution tomographic image of each nuclide identified in the time-correlated gamma-ray spectrum. By detecting only neutrons that have the proper flight-time to be uncollided, the APSTNG can also form the basis for a compact fast-neutron transmission imaging system with no collimator that can be used along with or instead of the emission imaging system.

2. APSTNG System Characteristics

The diagnostic probe which Argonne has recently been supporting was developed by the Advanced Systems Division of Nuclear Diagnostic Systems (NDS).[2,3] Its operation can be understood from Fig. 1. 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. 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 TAC's (time-to-amplitude converters), digitized by an ADC (analog-to-digital converter), and recorded.

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 scanning the alpha detector horizontally and vertically, or 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.

Figure 2 illustrates the electronics and information flow for a basic multipixel system containing a 2D position-sensitive alpha detector. In Fig. 2, the vectors involved in the reaction location are "Rt", "Rd", and "Rs". The transverse "X" and "Y" coordinates of "Rt" are digitized and stored in the PC computer, along with "TOF" (time-of-flight = "t" - "T") and gamma-ray energy "Eg". The PC controls the experiment, calculates positions, and displays data and images.

2.1 DIFFERENT OPERATIONAL 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 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. Slow-neutron capture is not time-correlated with the alpha pulses, but provides nonimaging gamma-ray spectra that can aid nuclide identification. The use of non-correlated gamma-ray spectra is termed the CGRS (capture gamma-ray spectroscopy) mode. (If fissionable materials are present, neutron reaction detectors may be used to detect emitted fission neutrons).

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 is feasible. Transmission imaging (FNTI) can be done along with or instead of emissive reaction-density imaging (EGRIS).

2.2 SEALED-TUBE NEUTRON GENERATOR

The use of the associated-particle method in arms control treaty verification and contraband detection research and development is recent. However the application of this method to NDA for neutron inelastic scattering is not new, although it has been relatively undeveloped and confined to the laboratory because of the bulk, complexity, and reliability and maintenance problems of the accelerator equipment previously required. The replacement of the accelerator in this neutron diagnostic probe system by the sealed-tube APSTNG brings new flexibility to the method and allows it to become a tool for field use. The state-of-the-art APSTNG was developed by the Advanced Systems Division of NDS, after considerable experience.

As diagrammed in Fig. 3, a Penning ion source inside the APSTNG emits a continuous mixed beam of deuterium and tritium ions that is accelerated and focused on a small spot on the target, tritiating the target and producing neutrons and alpha particles. A getter controls the mixture of deuterium and tritium. The single-pixel alpha detector consists of a ZnS screen inside the tube, with a photomultiplier outside. (In the case of a multipixel 2D alpha detector, the photomultiplier can be replaced by a microchannel plate and resistive anode readout.) The APSTNG is an inexpensive small sealed module with low-bulk support equipment. It has a long mean-time-between-failures (around 2000 hours at 10^6 n/s or 200 hours at 10^7 n/s), is easily replaced (allowing simple field operation), presents low radiation exposure, and the sealed-tube design prevents tritium contamination.

APSTNG tubes are commercially available from NDS. The operating history of each APSTNG made by NDS has been maintained and is available. Initial maximum output of a typical APSTNG is around 3×10^7 n/s, but the maximum output soon falls to about the level of 10^7 n/s. Eighteen ceramic tubes have been made, fourteen of which passed quality control tests (a 78% yield). Eleven of these tubes were put in operation (three are in stock), of which six are still operating. Three tubes failed in normal operation; two were failed by continuous operation at excessive voltage. For the nine tubes which were operated normally, the average lifetime output is 7.35×10^{12} neutrons, if end of life is defined as being reached when the maximum output becomes less than 10^6 n/s.

3. APSTNG Applications

APSTNG technology has the capabilities for identification and 3D imaging of many individual nuclides and compounds, with flexible positioning of reaction detectors with respect to the neutron source (on the same side, perpendicular, or opposite side), as well as capability for fast-neutron transmission imaging. The source and emitted radiation are high-energy and penetrate highly absorbing objects. Proof-of-concept laboratory experiments have been successfully done for a number of applications: nuclear warhead detection, chemical ordnance identification, explosive detection and identification, contraband drug detection, uranium borehole logging, corrodent detection on steam-turbine blades, kerogen analysis of shale,[4] on-line assay of coals, and bulk soil remediation of radwaste and pollutants.[5]

3.1 TREATY VERIFICATION ROLES

For the proof-of-concept experiment on nuclear warhead detection in the FNTI mode, a mockup was made consisting of two hollow Pb spheres (20 cm OD, 10 cm ID) separated by ~ 3 cm (simulating SNM) embedded in dry sand (simulating high explosive or propellant) in a 50-cm diameter container. Measurements were taken using a plastic neutron detector and a single-pixel APSTNG, with the alpha detector masked so the pixel diameter was ~ 2.5 cm at the mockup position. The experiment setup is shown in Fig. 4 (the detector is located far enough from the interrogated object that forward-peaked scattering is negligible). The resulting signal transmission given in Fig. 5 shows the Pb spheres, their hollow nature, and the sand. The transmission profile data is in good agreement with a simple convolution of the pixel diameter and the expected attenuated projection of the mockup from known cross-section data for 14-MeV neutrons, indicating that for a 2D detector or hodoscope array, image spatial resolution will be limited by spatial resolution of the neutron detection system, rather than by flight-time resolution. These experiments and other experiments on transmission through thick depleted-U blocks demonstrate high penetration but adequate contrast, for treaty verification roles.

For the proof-of-concept experiment on nuclear warhead detection in the EGRIS mode, a mockup was made consisting of two 12.7 cm x 15.2 cm x 15.2 cm solid blocks of depleted U (simulating SNM) separated by 10.2 cm and surrounded on all sides by 15.2 cm NH_4HCO_3 (simulating conventional high explosive or propellant). Measurements were taken using an APSTNG containing a single-pixel alpha-particle detector and a 15.2 cm x 35.6 cm NaI gamma-ray detector. When the two blocks were aligned along the axis of the APSTNG correlated neutron beam, the overall energy spectrum shown in Fig. 6 was obtained. The energy bands shown were used to separately image uranium (fission gamma-ray spectrum) and ammonium bicarbonate (oxygen inelastic scattering lines) along the beam axis, leading to the results shown in Fig. 7. Although the signal amplitudes are smeared out by the finite system time resolution (caused primarily by the large NaI detector), the U blocks and ammonium bicarbonate regions are correctly identified. The signal from the block farthest from the APSTNG is readily apparent, despite substantial attenuation by the closest block and by the ammonium bicarbonate.

An investigation of chemical ordnance configurations, gamma-ray energies and cross-sections for neutron inelastic scattering, and element structure of CW agents and explosives indicates that the EGRIS mode should be capable of detecting and distinguishing nearly all CW agents, by using ratios of element gamma-line intensities. For a very early version of an EGRIS system, measurements were taken of 155-mm shells filled to the proper levels with CW simulants. Resulting spectra (yielding oxygen, phosphorus, and sulfur peaks of various magnitudes) are shown in Figs. 8-10 for VX, GB, and mustard simulants, with shell spectrum subtracted. The spectra are different enough to be distinguished by eye, without quantitative analysis.

3.2 DETECTION OF EXPLOSIVES AND DRUGS

Based on data from proof-of-concept APSTNG laboratory experiments on explosives and drugs, it is interesting to estimate the measurement times required for detection of contraband items using a fielded APSTNG system, in this case, one having a multipixel alpha-particle detector and six relatively large gamma-ray detectors. A plot of the ratio of nitrogen to oxygen to that of carbon to oxygen is useful for differentiating contraband drugs or explosives from ordinary items expected to be seen, such as Fig. 11. Materials that would normally appear in luggage or foodstuffs are separated from explosives (open squares) and cocaine.

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 definitely identified as an explosive. In fact, even a 4-second measurement would distinguish the 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 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 why would plastic appear in fish?).

3.3 USE OF DIFFERENT MODES

For the most part, the proof-of-concept experiments have been conducted using only time-correlated, or imaging, reaction gamma-ray spectra (from neutron inelastic scattering for nearly all applications), in the IGRIS mode. Analysis of the soil remediation experiments included the uncorrelated gamma-ray spectrum, called the CGRS mode. The FNTI mode has so far not been much used, but it may find use in the future for simultaneous mode measurements, to correct IGRIS images for neutron attenuation or for multimode image analysis, or in a separate system for imaging extended or highly absorbing objects.

The IGRIS mode is not very suitable for imaging extended objects much over a meter or so in all three dimensions or highly absorbing objects. For large objects, the double solid angle reduction (source-to-object and object-to-detector) substantially reduces signal count rate. For large objects, accidental counts become limiting, and the raw images exhibit amplitude reductions and increased fluctuations in regions where neutron or gamma-ray attenuation are significant.

In the FNTI mode, the signal is proportional to the source-to-detector solid angle alone. It may be feasible to combine FNTI hodoscope arrays with GRTI (gamma-ray transmission imaging) hodoscope arrays so as to obtain Z-sensitive images of large volumes, such as a truck or cargo container, by employing the differences in attenuation Z-dependence (tomographic imaging may be necessary to do this). In some applications, this combination of FNTI and GRTI may allow determination of suspicious regions inside the volume, with a low false alarm rate. Then a two-stage interrogation process could be applied, in which the suspicious region would be offloaded and examined using a separate system based on the EGRIS mode.

4. Conclusions

The proof-of-concept experiments for nuclear warhead detection show a potential for high-confidence measurements in treaty verification applications for detection, counting, and dismantlement of nuclear weapons. The proof-of-concept experiments for CW agents indicate a means other than chemical sampling is available and should be effective for verification of these agents, a method that requires no opening of sealed containers, penetrates intervening materials, and is nonintrusive regarding sensitive molecular bond information. The treaty verification instrumentation criteria of transportability, reliability, easy operation and maintenance, personnel safety, and relatively low cost, would be met in these applications. Intrusion can be limited by limiting allowed output, measurement time, or time, space, or energy resolution, or by encryption of data. Combining hodoscope technology with APSTNG technology opens avenues to a wide range of applications, not only for treaty verification, but also for detection of contraband drugs and explosives and remediation of radwaste and pollutants.

However there are some limitations in the IGRIS mode of the present APSTNG system that can be significant for a number of applications, even for interrogation of volumes that are not large. Presently attainable depth resolution is limited, for a small gamma-ray detector, to about 5 cm (because the system has an overall time resolution of ~ 1 ns and a 14-MeV neutron travels 5 cm in 1 ns), and measurement times can be rather long to obtain sufficient gamma-ray counts. The gamma-ray signal count rate is limited by reaction cross-sections, solid angles subtended by the alpha detector and gamma-ray detectors, gamma-ray detector efficiency, and neutron source strength, but usable source strength is limited by detector accidental counts and pileup.

A gamma-ray hodoscope detector array can be used to substantially increase the signal count rate to lower measurement times, when necessary. However, small detectors will have a small energy-peak efficiency for the high-energy gamma-rays detected, while large detectors of the standard form will degrade the time resolution (and thus the depth resolution) substantially. Detector configurations are being investigated that will substantially improve the signal count statistics, while retaining as much time and energy resolution as possible.

5. Acknowledgments

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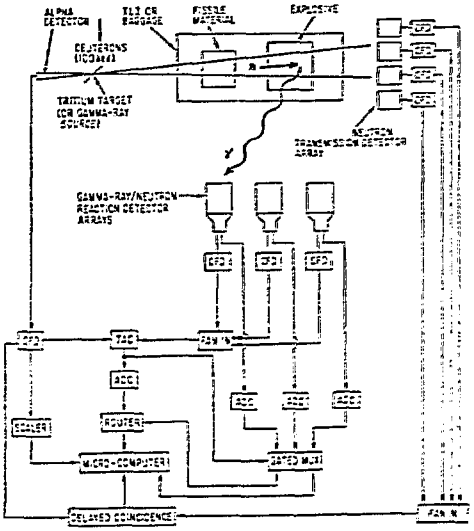

Fig. 1. Schematic layout of APSTNG-based interrogation system.

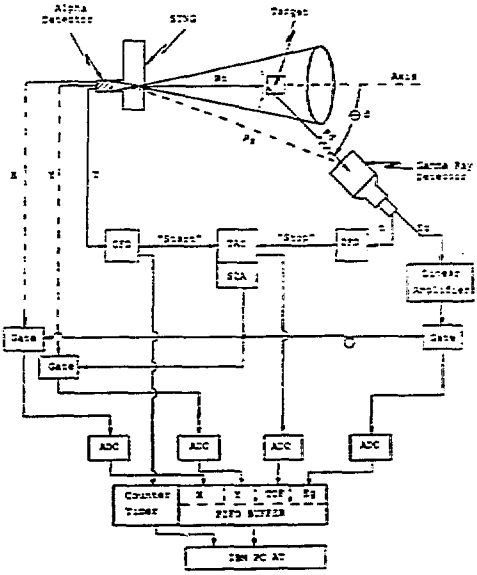

Fig. 2. Electronics and information flow for basic multipixel APSTNG-based system with 2D position-sensitive alpha-particle detector.

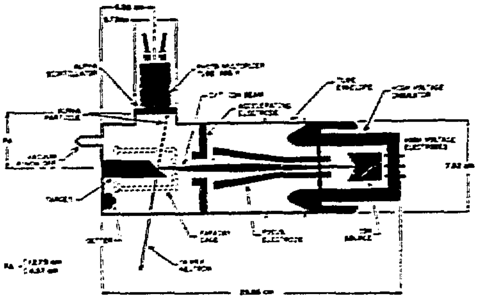

Fig. 3. Diagram of interior of APSTNG (Associated-Particle Sealed-Tube Neutron Generator).

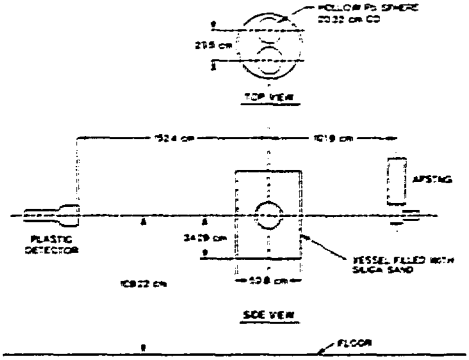

Fig. 4. Experiment setup for FNTI measurement of Pb spheres in sand, perpendicular to neutron beam.

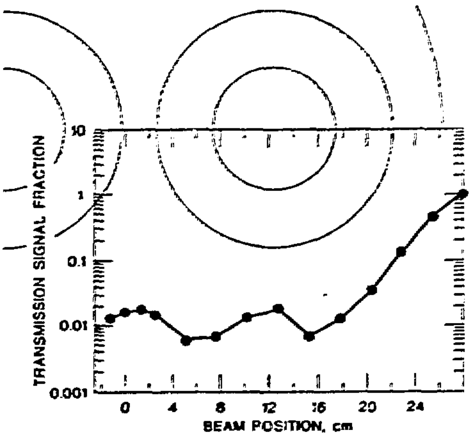

Fig. 5. FNTI signal transmission for hollow Pb spheres embedded in dry sand, perpendicular to neutron beam.

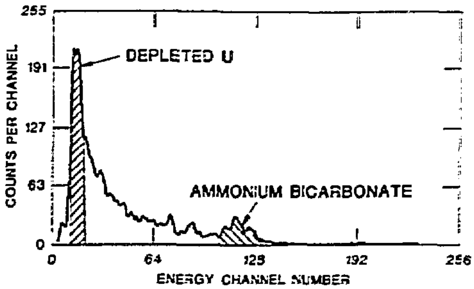

Fig. 6. Energy bands used for selection of EGRIS events primarily due to depleted-U and ammonium bicarbonate.

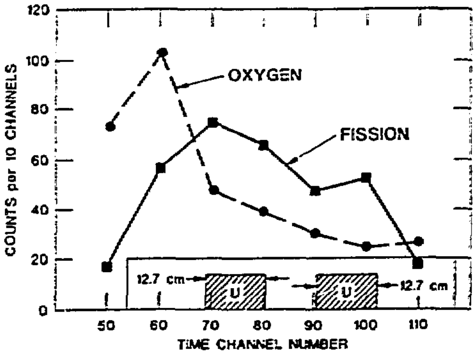

Fig. 7. EGRIS energy band signals for depleted-U and ammonium bicarbonate vs. flight time.

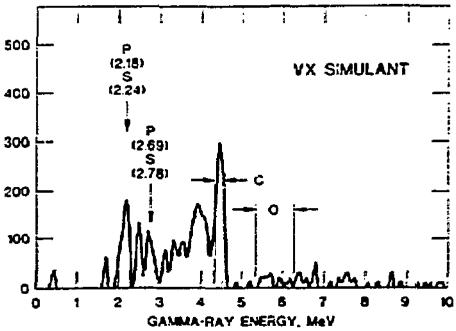

Fig. 8. Inelastic gamma-ray spectrum of 155-mm shell filled with bensulfide-xylene (54%) as VX simulant.

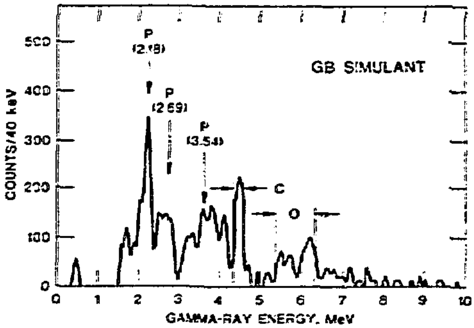

Fig. 9. Inelastic gamma-ray spectrum of 155-mm shell filled with dimethyl methyl phosphonate as GB simulant.

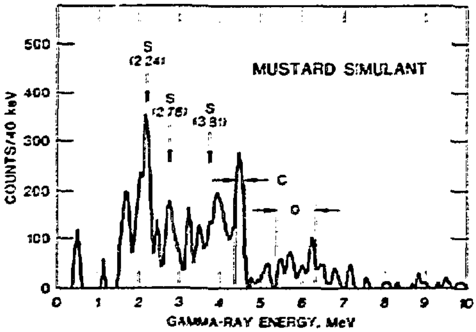

Fig. 10. Inelastic gamma-ray spectrum of 155-mm shell filled with thiodiglycol as mustard simulant.

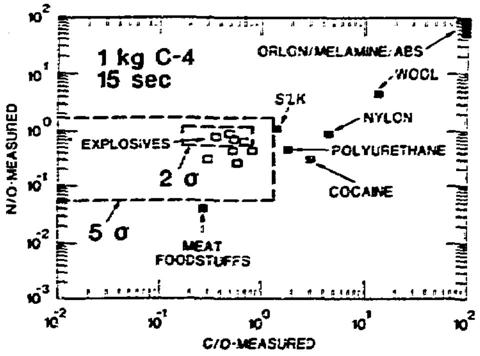

Fig. 11. APSTNG N/O vs. C/O for 15-second measurement of 1 kg C-4 explosive. Large box envelops 5-sigma statistics and small box envelops 2-sigma statistics.

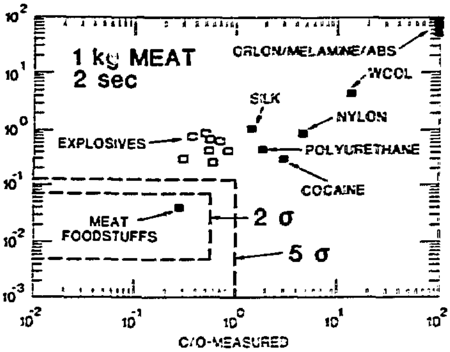

Fig. 12. APSTNG N/O vs. C/O for 2-second measurement of 1 kg of meat. Large box envelops 5-sigma statistics and small box envelops 2-sigma statistics.

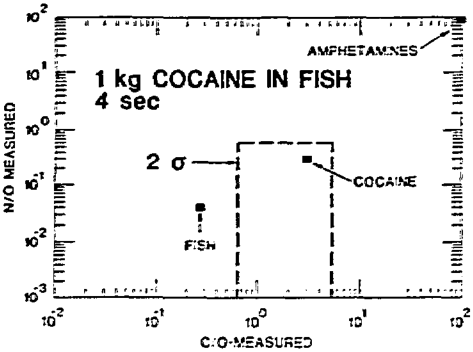

Fig. 13. APSTNG N/O vs. C/O for 4-second measurement of 1 kg of cocaine in fish. Box envelops 2-sigma statistics.