ADVANCES IN ASSOCIATED-PARTICLE SEALED-TUBE NEUTRON PROBE DIAGNOSTICS FOR SUBSTANCE DETECTION

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Advances in associated-particle neutron probe diagnostics for substance detection*

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ABSTRACT

The development and investigation of a small associated-particle sealed-tube neutron generator (APSTNG) show potential to allow the associated-particle diagnostic method to be moved out of the laboratory into field applications. The APSTNG interrogates the inspected object with 14-MeV neutrons generated from the deuterium-tritium reaction and detects the alpha-particle associated with each neutron inside a cone encompassing the region of interest. Gamma-ray spectra of resulting neutron reactions identify many nuclides. Flight-times determined from detection times of the gamma-rays and alpha-particles can yield a separate coarse tomographic image of each identified nuclide, from a single orientation. Chemical substances are identified by comparing relative spectral line intensities with ratios of elements in reference compounds. The high-energy neutrons and gamma-rays penetrate large objects and dense materials. Generally no collimators or radiation shielding are needed. Proof-of-concept laboratory experiments have been successfully performed for simulated nuclear, chemical warfare, and conventional munitions. Most recently, inspection applications have been investigated for radioactive waste characterization, presence of cocaine in propane tanks, and uranium and plutonium smuggling. Based on lessons learned with the present APSTNG system, an advanced APSTNG tube (along with improved high voltage supply and control units) is being designed and fabricated that will be transportable and rugged, yield a substantial neutron output increase, and provide sufficiently improved lifetime to allow operation at more than an order of magnitude increase in neutron flux.

1. INTRODUCTION

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 remediation 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 and detects the alpha-particle associated with each neutron and energy and flight-time spectra from resultant gamma-rays.

Investigations of applications for verification of chemical and nuclear weapons[1], detection of explosives and drugs[2,3], and extension to large interrogation volumes[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; these most recent studies will be described in the following sections. Then design, development, and fabrication of the new advanced APSTNG

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tube (along with the improved high voltage supply and control unit) will be discussed. But first, in this section, operational aspects of the present system and its components will be described, including detection modes and the sealed-tube neutron generator.

1.1 Principles of operation

The neutron diagnostic probe which Argonne has been using was developed by the Advanced Systems Division of 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, that could contain explosives or drugs or CW or fissile 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. A single specially configured gamma-ray detector is employed in the current ANL APSTNG system. A NaI(Tl) crystal 10.16 cm × 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.) 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 at least 0.5 barn or so for 14-MeV neutrons (predominantly inelastic scattering reactions). For gamma-rays above 1 MeV or so, background is greatly reduced since background counts can only be accumulated during the nanosecond-range correlation interval, yielding a high signal-to-background ratio. Because neutrons are emitted isotropically, the source and emission detection systems can be located arbitrarily, on the same side when access is restricted. 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. The 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 multichannel analyzer board inside the PC. The thermal neutron capture cross section is small for most nuclides, but is large for a few 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 used to map and position the neutron correlation cone but so far has not been used for interrogation.

1.3 Sealed-tube neutron generator

As diagrammed in Fig. 3, 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, tritiating the target and producing neutrons and alpha particles. (The small spot is necessary to obtain good spatial resolution.) A zirconia 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 μA to get 10^6 n/s) is controlled by varying the getter heating current. All operating voltages and currents are furnished 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. (In the case of a multipixel 2D alpha detector, the photomultiplier can be replaced by a microchannel plate and matrix anode readout, as shown in Fig. 3.) 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 3×10^7 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. 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, in the form of beads of density 0.576 g/cm^3, yielding a carbon density of 0.492 g/cm^3. This compares favorably with propane, with density 0.501 g/cm^3 and carbon density of 0.410 g/cm^3. Table IV describes candidate surrogates for cocaine and cocaine hydrochloride made from commercially available plastic feedstocks and carbon (C), including polyacrylonitrile (PAN), poly(1,4 butylene terephthalate) (PBT), polymethyl methacrylate (PMMA), polyethylene (PE) and polyvinyl chloride (PVC). The presence of black carbon granules in two of the candidates in Table IV has the potential of being used for visual estimates of the mix uniformity, but only the candidate labeled "Cocaine Hcl (used)" was made.

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. As shown in Table IV (on the next page), the materials were then combined in ratios such that the final mixed product duplicated the relative elemental densities of cocaine hydrochloride. 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 "nuclear" simulant of cocaine Hcl.

The cocaine Hcl surrogate in its cylindrical bottle has a net density of 0.709 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 ~ 4×10^6 n/s.

Table IV. Candidate Surrogate Mixtures of Cocaine [$C_{17}H_{21}NO_4$] and Cocaine Hydrochloride [$C_{17}H_{22}ClNO_4$]

Drug Simulated	Component	Relative No. Moles	Per Cent by Weight	Atoms Contributed C	H	Cl	N	O
Cocaine	PMMA	2	66.0	10	16	---	---	4
(suggested)	PAN	1	17.5	3	3	---	1	---
	PE	0.5	4.6	1	2	---	---	---
	C	3	11.9	3	---	---	---	---
	Total Mix		100.0	17	21	---	1	4
Cocaine	PAN	1	15.6	3	3	---	1	---
Hcl	PVC	1	18.4	2	3	1	---	---
(used)	PMMA	1	29.5	5	8	---	---	2
	PBT	0.5	32.4	6	6	---	---	2
	PE	0.5	4.1	1	2	---	---	---
	Total Mix		100.0	17	22	1	1	4
Cocaine	PMMA	2	58.9	10	16	---	---	4
Hcl	PAN	1	15.6	3	3	---	1	---
(suggested)	PVC	1	18.4	2	3	1	---	---
	C	2	7.1	2	---	---	---	---
	Total Mix		100.0	17	22	1	1	4

Figure 3 shows the result of subtracting the normalized energy spectrum of simulated propane from the energy spectrum of simulated cocaine hydrochloride. Note that this subtraction results in approximately zero net counts in the carbon peaks, providing an independent 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 especially prominent. Counting time for each of these two measurements on relatively small samples was 60 minutes. A much smaller measurement time than this would be required to determine the presence of cocaine Hcl with sufficient statistical significance.

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 ENIS mode would appear to have an advantage over the EGRIS mode, since the fission neutrons, unlike the fission gamma rays, do not have to be distinguished from spectra caused by other reactions. However the fission neutrons have a wide range of velocities and do not lose much energy in scattering, which cause substantial degradation in spatial resolution and difficulty in distinguishing the fission neutrons from incident APSTNG neutrons; these effects are exacerbated by the low energy resolution and high gamma-ray sensitivity of

typical fast-response fast-neutron detectors. Experiments performed by NDS during the initial advanced concepts stage with plastic neutron detectors and no spectrum unfolding and no collimators indicated that the EGRIS mode with fission gamma-ray detection was more promising.

Approximately 7 prompt gamma rays, with a total energy of about 7 MeV, are emitted, on average, per U-235 fission. The prompt fission gamma-ray spectrum peaks below 1 MeV, and then decreases approximately exponentially with increasing energy, up to about 7 MeV. The lowest energy gammas in this spectrum are undetectable against background and the highest energy gammas are too few to detect. In the 1-3 MeV detectable range, ~ 2.4 gamma rays are emitted per fission. The U-238 cross section for fission by 14-MeV neutrons is ~ 1 barn, about one-half that for U-235. Figure 4 shows the measured EGRIS prompt gamma-ray spectrum from APSTNG-induced fission in U-238. The U-238 object was a rectangular parallelopiped, 12.7 cm deep, 7.6 cm wide and 7.6 cm high. 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.

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 cm × 40 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 ~ 10^8 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. So the APSTNG system could provide a rather complete characterization of the object without opening the package.

The measurement times estimated for the heavily shielded case include aapproximate allowances for the difficulty in distinguishing the continuous fission gamma-ray spectrum from the accidental coincidence background and from spectra of other materials. We have not included potential improvements in sensitivity by optimizing analysis software and spectrum recognition algorithms, and by employing statistical criteria specific to types of interrogated objects expected. APSTNG hardware options to enhance fission signature recognition include use of coincident fission gamma-ray counts in the gamma detectors and use of neutron detectors in the ENIS mode.

The use of coincident fission gamma-ray counts to provide a more unique fission signature is based on the ~ 2.4 detectable gamma rays emitted per fission in the EGRIS spectrum, but it would result in a drastic reduction in the signal count rate and requires coincidence units to be procured and cabled between the gamma detectors in the array. The use of neutron detectors in the ENIS mode would require an array of large fast detectors capable of separating the neutron and gamma pulses and spectrum unfolding software to help distinguish fission neutrons from scattered APSTNG source neutrons. Source and/or detector collimation also might be needed. Additional study would be needed to determine whether either or both of these two hardware options would be appreciably more effective than detection of fission gammas by the standard EGRIS mode.

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 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 resulting 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.

Radwaste benchmark experiments were scheduled at the Idaho National Engineering Laboratory (INEL) Radioactive Waste Management Center (RWMC). It was decided to measure radwaste and calibration drums from the Stored Waste Examination Pilot Plant (SWEPP), locating the APSTNG system in the nearby TRUPACT building. The APSTNG visit had to be scheduled in accord with the requirements of the RWMC as an operating facility, and the visit was conducted the last week of September, 1994.

ANL selected the SWEPP heterogeneous mixed metals calibration drum configuration for preparatory laboratory measurements at ANL. This configuration is a 55-gallon steel drum of diameter 58 cm and length 88 cm containing column structures of plates and pipes of steel, lead, copper, and aluminum of various sizes. Three aluminum tubes of 40-mm i. d. positioned at different radii run the length of the drum and allow samples to be inserted inside the drum. A tube insert is provided that can carry a number of nominally 1-g high-purity Pu-239 Nuclear Accident Dosimeter (NAD) sources. Because of SWEPP operational needs for calibration drums, we designed and fabricated a calibration drum of our own, and used it in laboratory experiments at ANL in preparation for the INEL visit. The design of the ANL calibration drum is fashioned after the SWEPP heterogeneous mixed metals drum; however, the ANL drum has 84-mm i.d. sample tubes, to accommodate a wider range of sample items. The top third of this drum is a projection of the cross-sections of the SWEPP drum, from top to bottom, i.e., all of the column structures at various heights in the SWEPP drum are represented as shortened columns filling the top third of the ANL 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.

For the laboratory preparatory experiments, 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 6×10^6 n/s and the runs were 1 hour long. To avoid background from scattered neutrons in the energy spectrum for the EGRIS time regions of interest (ROIs)

caused by neutron response of the gamma detector, the gamma detector should be located far enough away from the interrogated object that any scattered neutrons arrive after the gamma rays. The larger the object, the greater this separation distance becomes, in this case 76 cm between the 55-gallon drum and the gamma detector, which would substantially decrease gamma-ray count rate. It was decided to move the gamma detector much closer to increase its count rate and accept some background from scattered neutrons, which provide a continuous contribution to the energy spectrum. The axis of the gamma-ray detector was parallel to the cone axis, and the side of the scintillator was 14 cm from the nearest drum edge, or 43 cm from the drum center. The scattered neutrons were found to distort the flight-time spectra enough that it was necessary to subtract a background flight-time spectrum (measured in a run in which the samples were absent), in order to easily identify the time ROIs.

A number of measurements were made with the samples were vertically centered in the top and bottom thirds of the drum. 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 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, shows a dominant fission gamma spectrum. Gamma-ray peaks from the other materials must be stripped off if the fission signature is to be normalized to estimate the amount of fissionable material present (and vice-versa). Also, the accidental count spectrum, and in this case the background spectrum from neutron scattering, must be subtracted.

The APSTNG neutron generator tube and its only available replacement were both apparently damaged by a faulty HV control unit during the preparatory laboratory experiments at ANL, so that neither tube could produce neutrons. However, the passive gamma-ray measurements of actual radwaste drums in the work schedule were performed, as planned, in the TRUPACT building at RWMC. Data were collected in the PGRS mode from the large double-ended NaI detector, generally 10 minute runs. Measurements were performed with Pu-239 NAD sources inserted in the center of a source holder that was placed inside the center tube in sludge calibration drum #1 (content code 1). The spectrum from 1 g of Pu-239 (a single NAD source) after subtraction of background, shows a sufficient Pu-239 signature that 1 g of Pu-239 is detectable in the middle of the concrete sludge drum.

Four 55-gallon radioactive waste drums of the concrete sludge category were measured in the PGRS mode. We did not have available results of detailed RWMC measurements, and did not know which of these drums, if any, contained plutonium. Three drums were found to contain Pu-239 and one drum was found to contain U-238, the latter isotope identified by the gamma-ray lines from daughter products. None of the drums measured was found to contain both Pu-239 and U-238. The gamma-ray spectra for the drums containing Pu-239 differed significantly only in the intensity of the Pu-239 peaks.

Shown in Fig. 6 is the spectrum obtained for sludge drum #RF074404275 (content code 4, 293 kg), 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 the 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. No evidence of the prominent but easily absorbed 186-kev gamma ray from U-235 decay was found in the spectrum for any of the drums measured. A very small U-235 peak, presumably from residual U-235 contained in the depleted uranium, has been seen in SWEPP radwaste drum spectra obtained with high-resolution HPGe detectors.

3. ADVANCED APSTNG SYSTEM

It is necessary to develop an APSTNG system of more advanced design, in order to meet field criteria for a number of important applications, as described above. 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. 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 is easily damaged by overheating). 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 ~ 10 times that for the NDS APSTNG tube, and the number of neutrons generated during the expected lifetime is ~ 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 ~ 7×10^6 per second at 10^8 n/s and ~ 7×10^7 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 sufficient luminosity, sufficient 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 ~ 365 nm will not be transmitted by fiber-optic windows). Many scintillators have been investigated, and ZnO(Ga) has been chosen as the best overall candidate. Ga doped ZnO is available in an appropriate grain size range and 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 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. 7) 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_6 insulating gas. The coupling unit will have a pressure gage and a pressure switch that provides a low-pressure interlock and panel

warning light. The new HV coupling unit will be fieldable, able to withstand mechanical shocks and vibrations and quick and easy to set up and take down.

The new HV supply and accelerator control system will protect the APSTNG neutron tube against voltage and current surges with limiting circuitry (unlike the NDS control unit, which has damaged tubes as it has aged), and an HV-kill switch will be provided that can be carried by an operator for shutdown in case any repetitive arcing occurs anywhere 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.

4. ACKNOWLEDGMENTS

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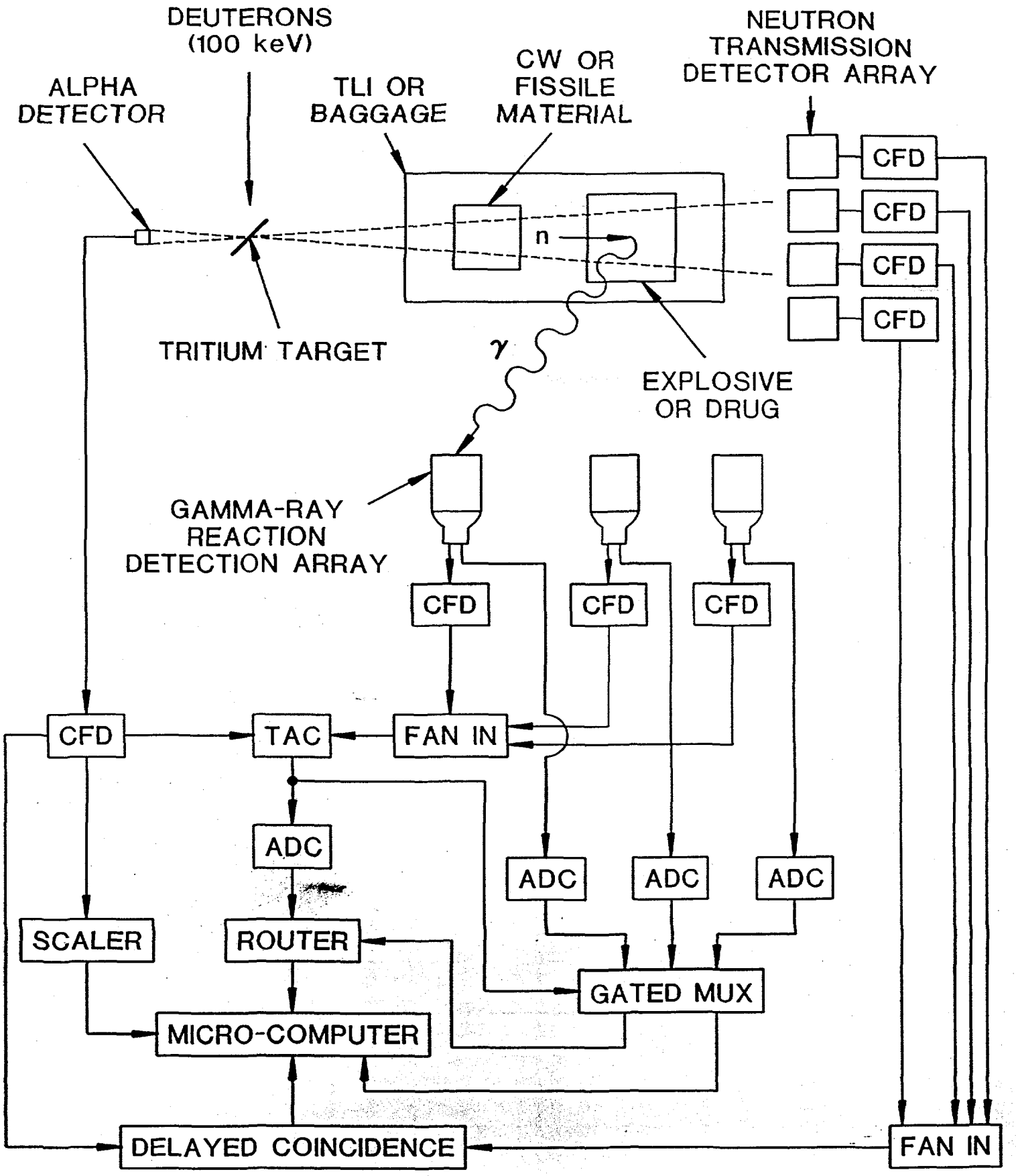

DEUTERONS
(100 keV)
ALPHA
DETECTOR
TLI OR
BAGGAGE
CW OR
FISSILE
MATERIAL
NEUTRON
TRANSMISSION
DETECTOR ARRAY
CFD
CFD
CFD
CFD
n
TRITIUM TARGET
γ
EXPLOSIVE
OR DRUG
GAMMA-RAY
REACTION
DETECTION ARRAY
CFD
CFD
CFD
CFD
TAC
FAN IN
ADC
ADC
ADC
ADC
SCALER
ROUTER
GATED MUX
MICRO-COMPUTER
DELAYED COINCIDENCE
FAN IN

07519

FIG. 1

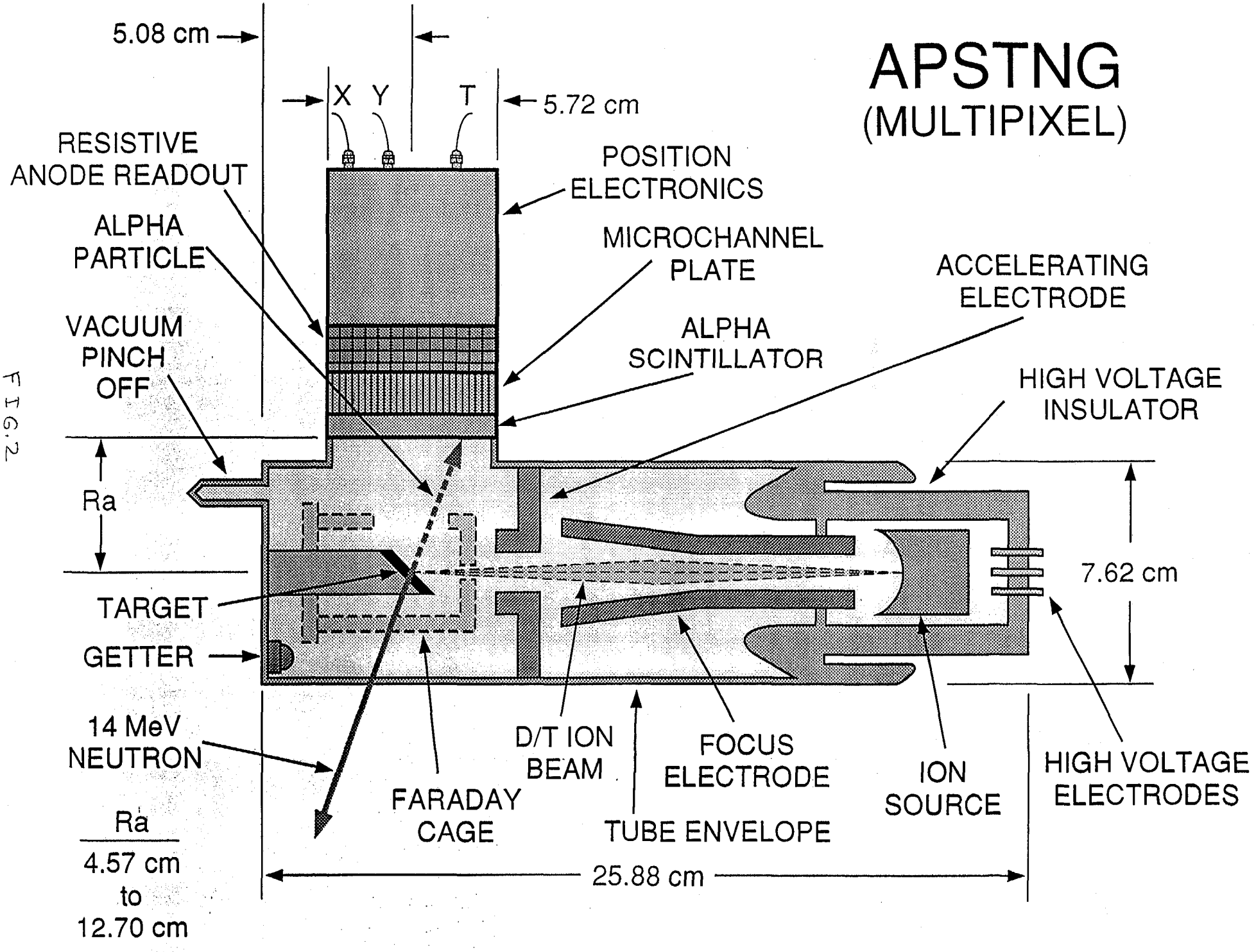

APSTNG
(MULTIPIXEL)
5.08 cm
X
Y
T
5.72 cm
RESISTIVE
ANODE READOUT
POSITION
ELECTRONICS
ALPHA
PARTICLE
MICROCHANNEL
PLATE
ACCELERATING
ELECTRODE
VACUUM
PINCH
OFF
ALPHA
SCINTILLATOR
HIGH VOLTAGE
INSULATOR
Ra
7.62 cm
TARGET
GETTER
14 MeV
NEUTRON
D/T ION
BEAM
FOCUS
ELECTRODE
ION
SOURCE
HIGH VOLTAGE
ELECTRODES
FARADAY
CAGE
TUBE ENVELOPE
Ra
4.57 cm
to
12.70 cm
25.88 cm

FIG. 2

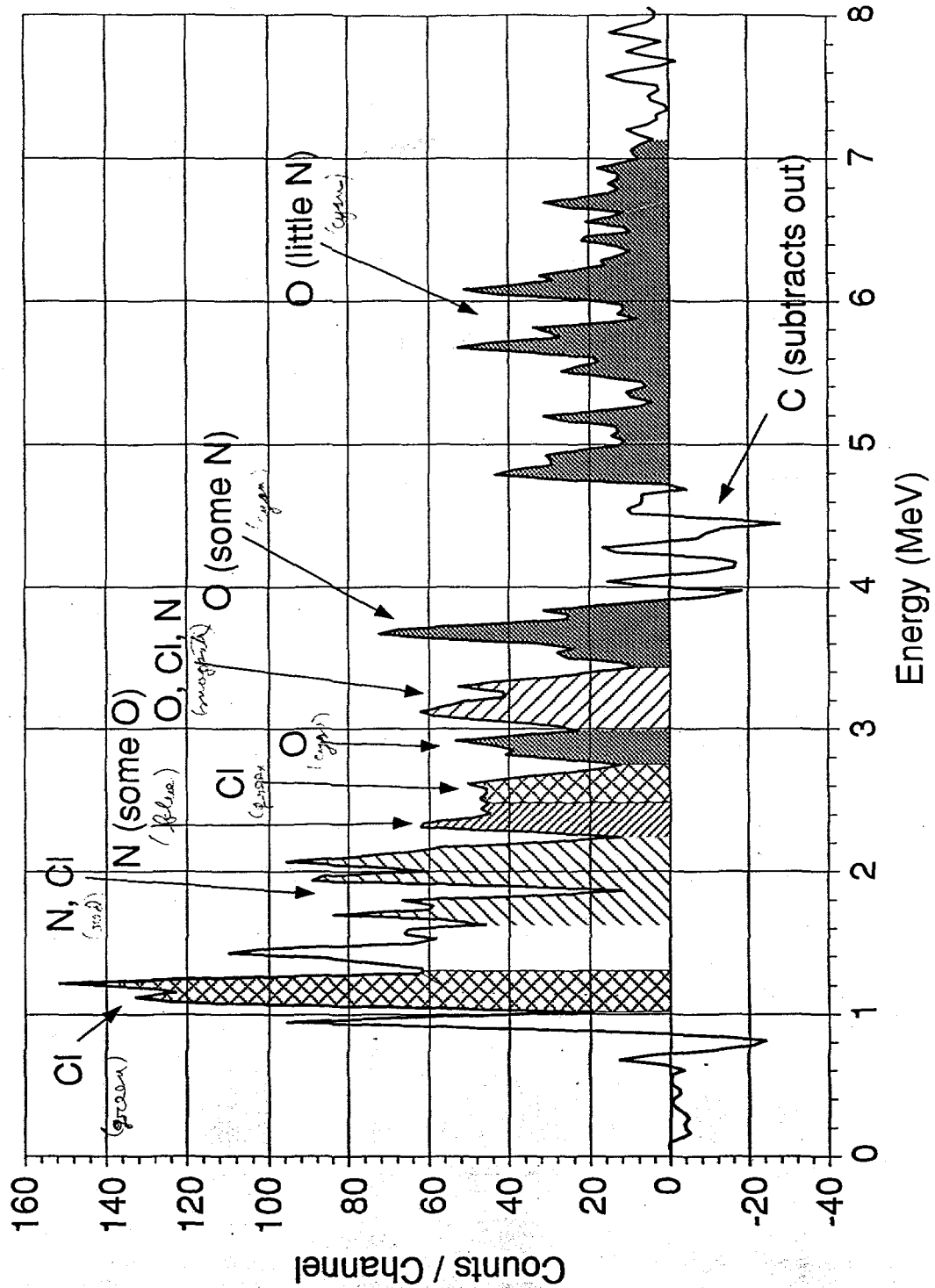

Cl
N, Cl
N (some O)
O, Cl, N
Cl
O
O (some N)
O (little N)
C (subtracts out)
Counts / Channel
Energy (MeV)
160
140
120
100
80
60
40
20
0
-20
-40
0
1
2
3
4
5
6
7
8

FIG. 3

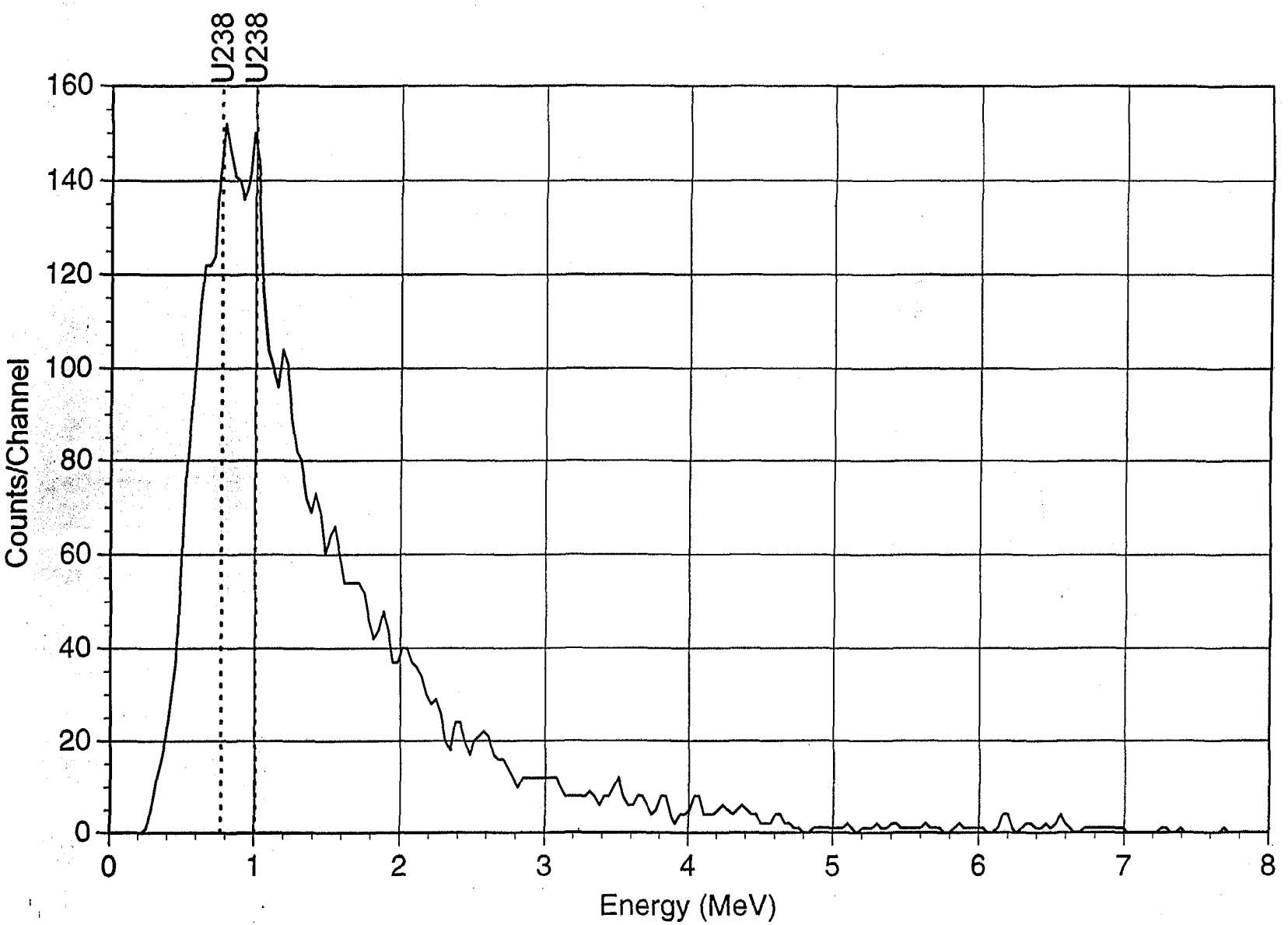

U238
U238
160
140
120
100
80
60
40
20
0
Counts/Channel
0
1
2
3
4
5
6
7
8
Energy (MeV)

FIG. 4

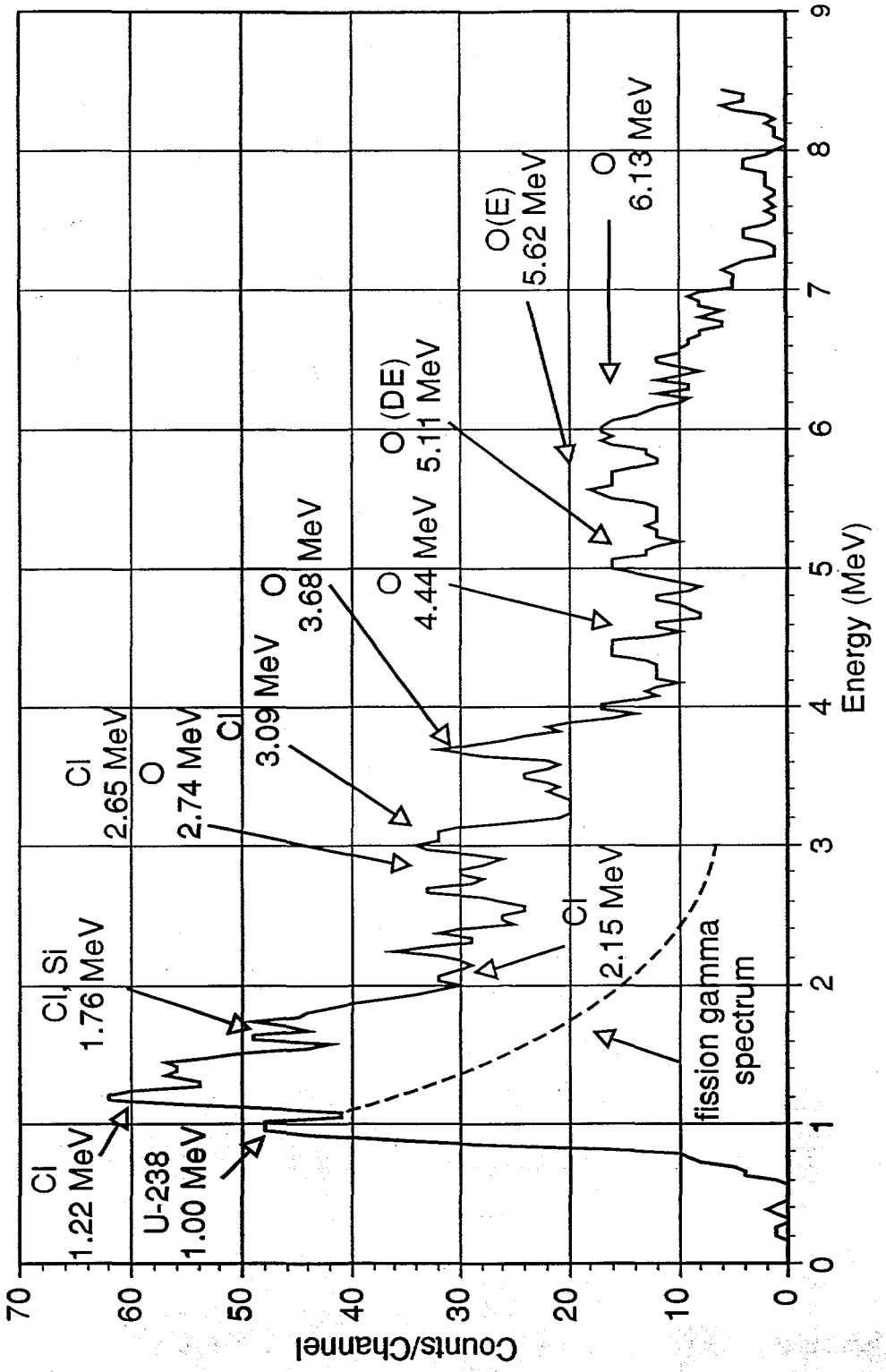

Counts/Channel
Energy (MeV)
Cl
1.22 MeV
U-238
1.00 MeV
Cl, Si
1.76 MeV
Cl
2.65 MeV
O
2.74 MeV
Cl
3.09 MeV
O
3.68 MeV
O
4.44 MeV
O (DE)
5.11 MeV
O(E)
5.62 MeV
O
6.13 MeV
Cl
2.15 MeV
fission gamma
spectrum

FIG. 5

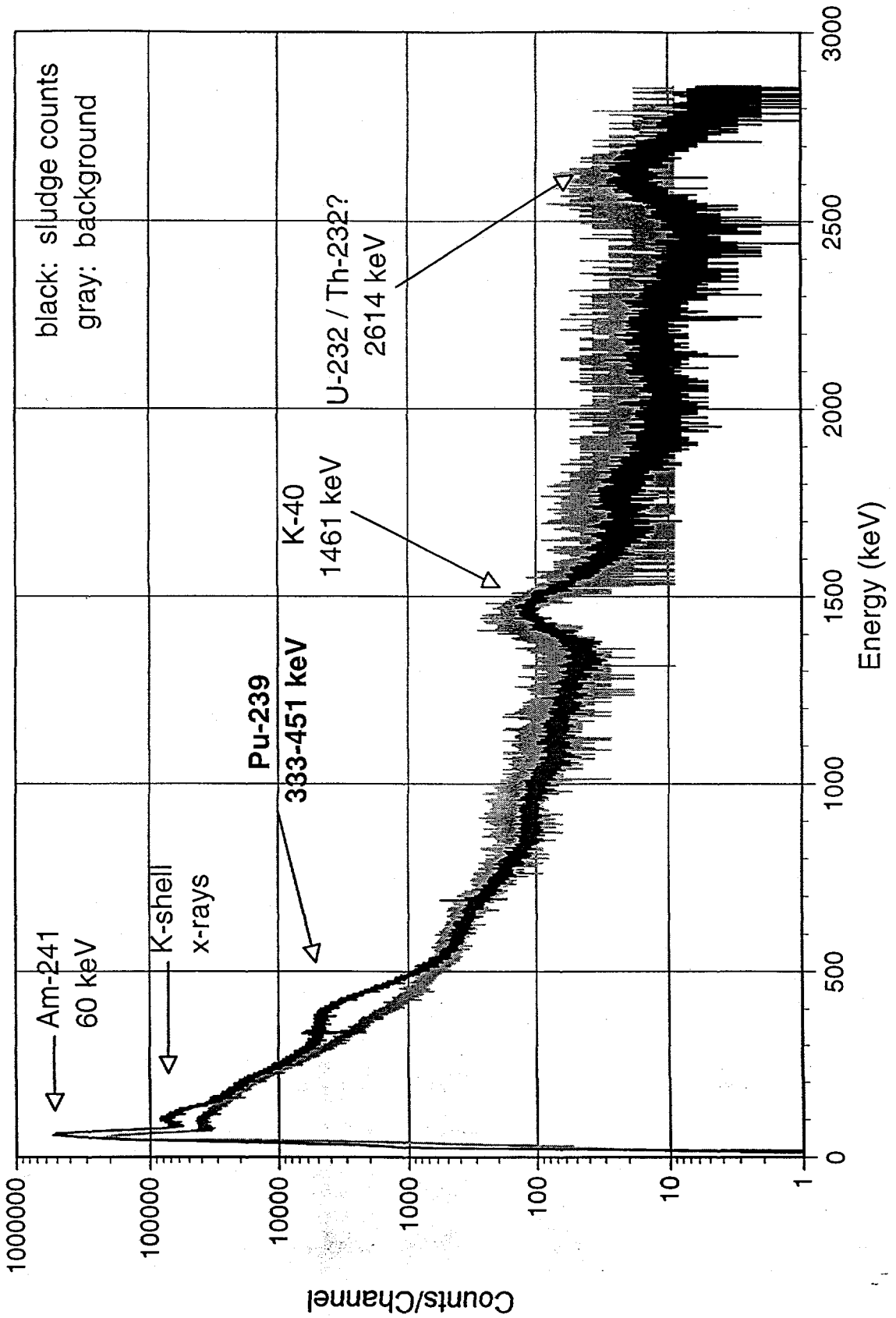
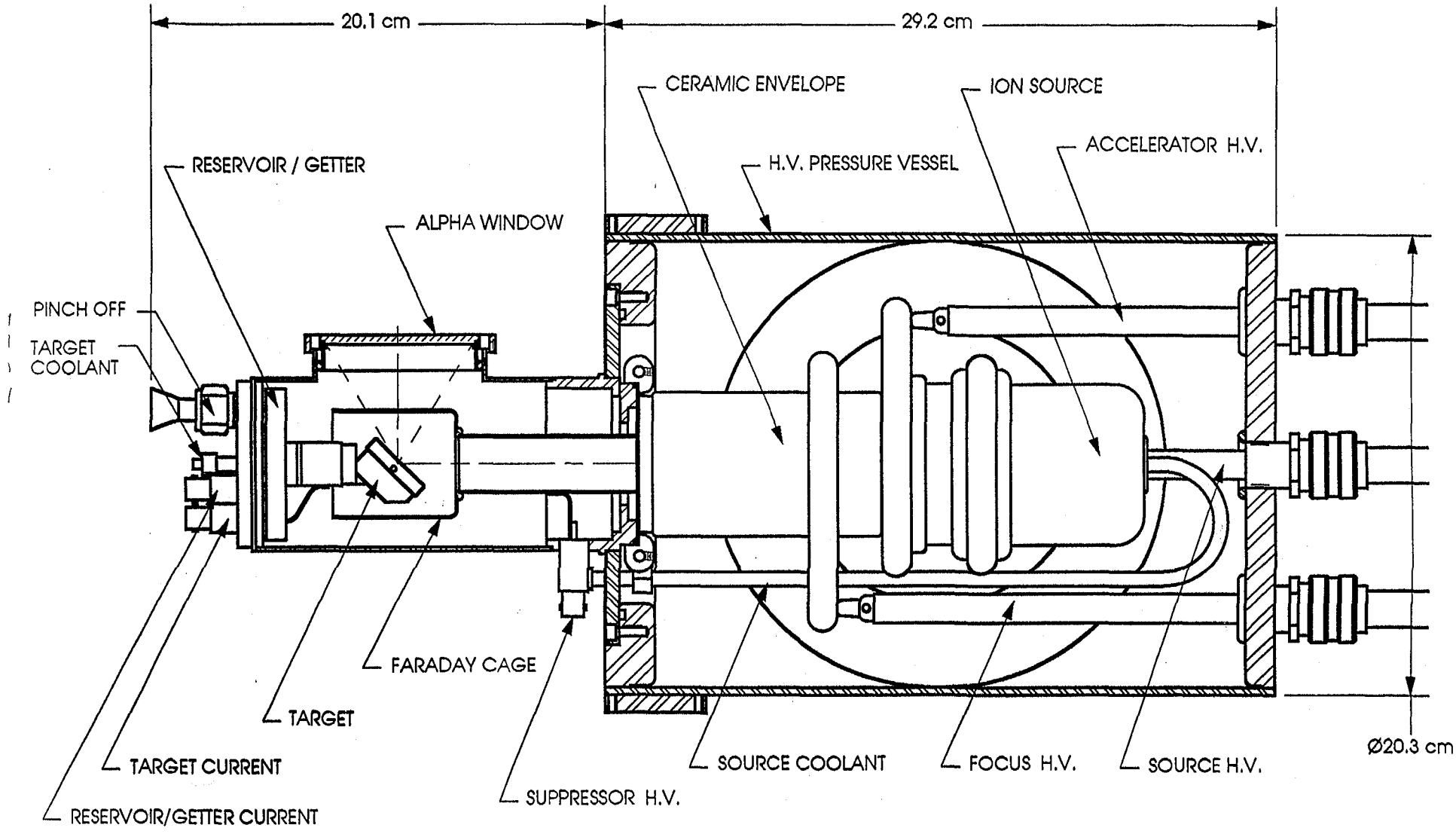

black: sludge counts
gray: background
Am-241
60 keV
K-shell
x-rays
Pu-239
333-451 keV
K-40
1461 keV
U-232 / Th-232?
2614 keV
Counts/Channel
Energy (keV)
1000000
100000
10000
1000
100
10
1
0
500
1000
1500
2000
2500
3000

FIG. 6



ACCELERATOR HEAD