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## WASTES CHARACTERIZATION USING APSTNG TECHNOLOGY

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# WASTE CHARACTERIZATION USING APSTNG TECHNOLOGY\*

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## ABSTRACT

The associated-particle sealed-tube neutron generator (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 from resulting neutron reactions inside the inspected volume identify fissionable materials and many nuclides. Flight-times determined from detection times of the gamma-rays and alpha-particles separate the prompt and delayed gamma-ray spectra and can yield coarse tomographic images from a single orientation. The high-energy neutrons and gamma-rays penetrate large objects and dense materials. The gamma-ray detector and neutron generator can be located on the same side of the interrogated object, so walls and other confined areas can be inspected, as well as sealed containers.

No collimators or radiation shielding are needed, the neutron generator is relatively simple and small, and commercial-grade electronics are employed. A complete system could be transported in an automotive van. Laboratory experiments and limited field tests indicate APSTNG technology could be useful in characterizing radioactive waste in drums, walls, soils, and processing systems, particularly for unknown or heterogeneous configurations that may significantly attenuate radiation. Also, toxic chemicals could be identified that would help classify the radwaste as to mixed waste content, and the ability to detect pockets of water may address criticality concerns.

## 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 special nuclear material and 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 (Rhodes et al., 1992a), detection of explosives and drugs (Rhodes and Peters, 1992b and 1992c), and extension to large interrogation volumes (Rhodes and Peters, 1992c; Rhodes et al., 1993) 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. Also, a new fieldable advanced APSTNG system (including neutron generator tube, high voltage supply, and control unit) has been designed and is being constructed.

Other NDA systems based on neutron interrogation and detection of gamma-ray spectra from resulting neutron interactions

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have also been recently proposed for examination of radioactive waste. Along with the APSTNG, these systems potentially may be used for a wide range of on-site applications in support of facility D&D (decontamination and decommissioning), including facility characterization prior to D&D; monitoring progress of D&D operations; characterization of the waste as it is collected, treated and/or separated, and packaged; characterization of clean or decontaminated material; and facility characterization at completion of D&D. The data collected by these other systems is similar to that collected by the APSTNG, although the APSTNG has some advantages in data collection and specific applications.

Therefore we propose to use the advanced APSTNG as a test bed for facility D&D, using laboratory mockups and limited field tests. In this paper, operation of the APSTNG system is reviewed, its application to radioactive waste characterization and use as a test bed for facility D&D are discussed, and development of the new advanced APSTNG system is described.

### APSTNG OPERATION

The neutron diagnostic probe which Argonne has been using was developed primarily by Nuclear Diagnostic Systems (NDS) (Gordon and Peters, 1990). 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.

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 photo-multipliers, 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.

### Detection Modes

The use of the time-correlated gamma-ray spectra is denoted the EGRIS (emissive gamma-ray imaging and spectroscopy) mode, which applies to fast-neutron reactions having subnanosecond response. 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). 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). Fissionable materials can be identified by the characteristic fission gamma-ray spectrum in the EGRIS mode, or neutron detectors may be used to detect emitted fission neutrons in the ENIS (emissive neutron imaging and spectroscopy) mode.

For gamma-rays above  $\sim 1$  MeV, background is greatly reduced in the EGRIS mode, 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.

Neutron capture reactions require slow neutrons, but the source neutrons scatter many times in slowing down, which requires milliseconds. Thus neutron capture events are not time-correlated with the alpha pulses on a nanosecond time scale, but provide 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 multichannel 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 significantly more complex than for EGRIS mode, with much more background. Neutron moderator material may be needed 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 for inspection 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 (Rhodes and Peters, 1992b; Rhodes et al., 1993).

### **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, tritiating 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 is limited by accidental

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

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.

### **CHARACTERIZATION OF RADIOACTIVE/TOXIC WASTE**

Fissionable isotopes are normally the most important materials to characterize in radioactive waste. 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 not penetrate actinide materials very well if substantial amounts of actinides are present.

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 half that for U-235. Figure 3 shows the EGRIS prompt gamma-ray spectrum from APSTNG-induced fission in U-238. EGRIS spectra measured for enriched U are similar to Fig. 3, 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.

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 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.

## APSTNG Radwaste Experiments

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  $\sim 6 \times 10^6$  n/s and the runs were 1 hour long.

Data from a representative measurement in the bottom concrete sludge section is shown in Fig. 4, 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<sub>2</sub> (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, shown in Fig. 5, corresponding to the position of the depleted U sample, shows a dominant fission gamma-ray spectrum.

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, as shown in Fig. 7, except that the Pu-239 peak group is replaced by the 766 and 1001 keV peaks from the Pa-234m daughter of U-238.

## The APSTNG as a Test Bed

In addition to the APSTNG, a number of other neutron interrogation technologies in which gamma-rays from induced neutron reactions are detected have been proposed for radwaste characterization. We plan to investigate our new APSTNG being constructed, for use in facility D&D operations. In addition to collecting data in the basic APSTNG modes, we also plan to operate our APSTNG under conditions that simulate key operational characteristics of these other technologies, so that the APSTNG would serve as a generic test bed. These technologies, designated as PFNA, N-SCAN<sup>TM</sup>, and PINS, are described briefly below.

Pentaleri and Gozani (1994) describe an approach to characterizing radwaste containers using a PFNA (pulsed fast-neutron analysis) system being developed for use at the INEL RWMC. This project draws upon technology developed for PFNA inspection of large objects, such as cargo trucks (Gozani, 1994). Neutrons in the 7-9 MeV range in pulses of nanosecond-range width are generated from a deuteron beam impinging on a pumped deuterium gas target, using a pelletron accelerator operating at several MV with a double-pulse buncher.

In the PFNA system, the neutrons are collimated into a beam that scans the inspection volume and gamma-ray spectra are collected from a large array of small NaI(Tl) detectors. Gamma-ray flight time relative to the neutron pulse indicates the reaction-site origin of the gamma-ray, allowing a coarse 3D image of materials identified in the inelastic scattering and fission gamma-ray spectra, similar to the APSTNG EGRIS mode. However, if there is significant neutron moderation, capture gamma spectra will be mixed in, unlike the separate simultaneous EGRIS and CGRS spectra obtained with the APSTNG. An array of transmission neutron detectors provides a coarse 2D radiograph, like the APSTNG FNTI mode.

Because the APSTNG system can measure many pixels simultaneously, it can require substantially less average neutron flux than a PFNA system for the same average signal rate, and because APSTNG neutron flux is continuous (not pulsed), it needs significantly less data-acquisition bandwidth for the same average neutron flux. The PFNA nanosecond-pulsed high-current high-voltage accelerator equipment is substantially more complex and massive than the APSTNG sealed tube equipment, and the PFNA collimator and shielding add further substantial size and mass. However, the PFNA system is not limited by accidental counts, unlike the APSTNG system, and is capable of substantially higher signal rates, suitable for high throughput in fixed portal applications.

The N-SCAN<sup>TM</sup> system is a commercial tractor outfitted with a neutron generator, gamma-ray detector, power supply, electronics, telemetry unit, and a collimator designed to inspect a truncated cone of contaminated soil. Data are relayed to a mobile data processing van, allowing survey of a field site along a planned grid. An HPGe detector is employed, the neutron generator is a conventional pulsed unit, and data acquisition and analysis are modifications of conventional well-logging methods (Ruddy et al., 1993). Pulse width is in the microsecond range, too long to be of use in flight-time analysis but short enough to differentiate inelastic scattering and fission gamma-rays from capture gammas, so as to yield separate spectra. The system has no direct imaging capability, but it is reported that approximate depth profiles can be calculated by algorithms making use of the differential attenuation of prompt gamma rays of different energies emitted from a single element.

The PINS (portable isotopic neutron spectrometer) is the simplest of the neutron interrogation systems considered. A radioactive Cf-252 fission neutron source is partially moderated, so that both prompt gamma-rays (primarily from inelastic scattering and fission) and capture gamma-rays are excited in the inspected object. These gammas are detected by a collimated HPGe detector that is shadow-shielded from the neutron source

and spectra are collected in a multichannel analyzer. Data are analyzed by a laptop computer that can be located up to 25 m from the detector. The PINS system is highly portable but cannot separate the prompt and capture spectra and has no imaging capability. It was originally developed for nondestructive field inspections to detect chemical warfare munitions and distinguish them from conventional munitions (Caffrey et al., 1992 and 1995).

The ANL APSTNG system is well suited to operation in a test-bed mode for these neutron interrogation technologies. The instrument can operate in the EGRIS mode to obtain prompt gamma-ray spectra from prompt reactions with fast neutrons, even though it uses a continuous source rather than a nanosecond pulsed source, like PFNA. It also can operate in the CGRS modes with or without moderator material, like PINS and PFNA. N-SCAN<sup>TM</sup> collects gamma-ray spectral data from both the prompt and uncorrelated categories. Both NaI and HPGe detectors are available for use with the APSTNG. Thus, APSTNG data can be provided directly relevant to PFNA, N-SCAN<sup>TM</sup>, and PINS.

Initial test-bed emphasis is expected to be on benchmarking, with simple laboratory mockups, effects of neutron scattering, absorption and thermalization; as well as effects of gamma-ray scattering and attenuation. The new ANL APSTNG also can be readily transported from the laboratory to D&D operations, as needed for application testing. Overall, these test-bed measurements should significantly facilitate the performance and interpretation of actual demonstration measurements with specific instruments. They also should assist in clarifying the potential problems associated with the materials and geometries of interest for practical applications. However, they are not a substitute for demonstration measurements with specific instruments, which also provide guidance on ease of use, portability, and maintainability.

## 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. 8, 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 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 needed for operation at  $10^9$  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 (pressure vessel) for the new neutron tube (see Fig. 9) 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  $\text{SF}_6$  insulating gas. The unit will have a pressure gage and a pressure switch that provides a low-pressure interlock and panel warning light.

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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#### REFERENCES

- Rhodes, E., Dickerman, C. E., DeVolpi, A., and Peters, C. W., 1992a, "APSTNG: Radiation Interrogation for Verification of Chemical and Nuclear Weapons", *IEEE Trans. Nucl. Science*, Vol. 39, pp. 1041-1045.
- Rhodes, E., and Peters, C. W., 1992b, "APSTNG: Neutron Interrogation for Detection of Explosives and Drugs and Nuclear and CW Materials", *SPIE Proceedings: Neutrons, X Rays, and Gamma Rays: Imaging Detectors, Material Characterization Techniques, and Applications (San Diego, California)*, Vol. 1737, pp. 160-170.
- Rhodes, E., and Peters, C. W., 1992c "APSTNG: Neutron Interrogation for Detection of Drugs and other Contraband", *Proc. Contraband and Cargo Inspection Technology International Symposium (Omni Shoreham Hotel, Washington, D. C.)*, pp. 37-44.
- Rhodes, E., Dickerman, C. E., and Peters, C. W., 1993, "Associated-Particle Sealed-Tube Neutron Probe for Characterization of Materials", *SPIE Proceedings: Substance Detection Systems (Innsbruck, Austria)*, Vol. 2092, pp. 288-300.
- Gordon, C. M., and Peters, C. W., 1990, "A Fast-neutron Probe for Tomography and Bulk Analysis", *Intl. J. Radiat. Appl. Instrum. Part A*, Vol. 41, pp. 1111-1116.
- Pentaleri, E., and Gozani, T., 1994, "Complete Characterization of Containerized Waste Using a PFNA-based Inspection System", *Nucl. Instrum. & Meth. in Phys. Research A*, Vol. 353, pp. 489-493.
- Gozani, T., 1994, "Novel Applications of Fast Neutron Interrogation Methods", *Nucl. Instrum. & Meth. in Phys. Research A*, Vol. 353, pp. 635-640.
- Ruddy, F. H., Congedo, T. V., Seidel, J. G., Gonzales J. L., and Weigle, D. H., 1993, "In-situ Characterization of Hazardous Contaminants Using Prompt Gamma Neutron Activation Analysis", Westinghouse Sci. & Tech. Report 93-7TD1-PGEXT-P1, presented at *Amer. Chem. Soc. 205th National Meeting, paper #78*.
- Caffrey, A. J., Cole, J. D., Gehrke, R. J., and Greenwood, R. C., 1992, "Chemical Warfare Agent and High Explosive Identification by Spectroscopy of Neutron-Induced Gamma Rays", *IEEE Trans. Nucl. Sci.*, Vol. 39, pp. 1422-1426.
- Caffrey, A. J., Gehrke, R. J., Greenwood, R. C., Hartwell, J. K., Krebs, K. M., and McLaughlin, G. D., 1995, "U. S. Army's Experience with the PINS Chemical Assay System", *Trans. Amer. Nucl. Soc.*, Vol. 72, pp. 125-126.

## FIGURE CAPTIONS

FIG. 1. SCHEMATIC LAYOUT OF "GENERAL PURPOSE" APSTNG-BASED INTERROGATION SYSTEM.

FIG. 2. CROSS-SECTION OF NDS APSTNG TUBE, WITH POSITION-SENSITIVE ALPHA-PARTICLE DETECTOR.

FIG. 3. EGRIS ENERGY SPECTRUM MEASURED FOR U-238 FISSION.

FIG. 4. CHLORINE AND DEPLETED URANIUM IN CONCRETE SLUDGE SECTION OF ANL RADWASTE CALIBRATION DRUM. EGRIS ENERGY SPECTRUM FOR FLIGHT-TIME CHANNELS 60-93.

FIG. 5. CHLORINE AND DEPLETED URANIUM IN CONCRETE SLUDGE SECTION OF ANL RADWASTE CALIBRATION DRUM. EGRIS ENERGY SPECTRUM FOR FLIGHT-TIME CHANNELS 93-119.

FIG. 6. PASSIVE GAMMA-RAY SPECTRUM OF RWMC RADWASTE SLUDGE DRUM RF074404275.

FIG. 7. PASSIVE GAMMA-RAY SPECTRUM OF RWMC RADWASTE SLUDGE DRUM RF74703133.

FIG. 8. CUT-AWAY DESIGN DRAWING OF NEW ADVANCED APSTNG CONSTRUCTED BY MF PHYSICS.

FIG. 9. DESIGN DRAWING OF ACCELERATOR HEAD FOR NEW ADVANCED APSTNG BY MF PHYSICS.





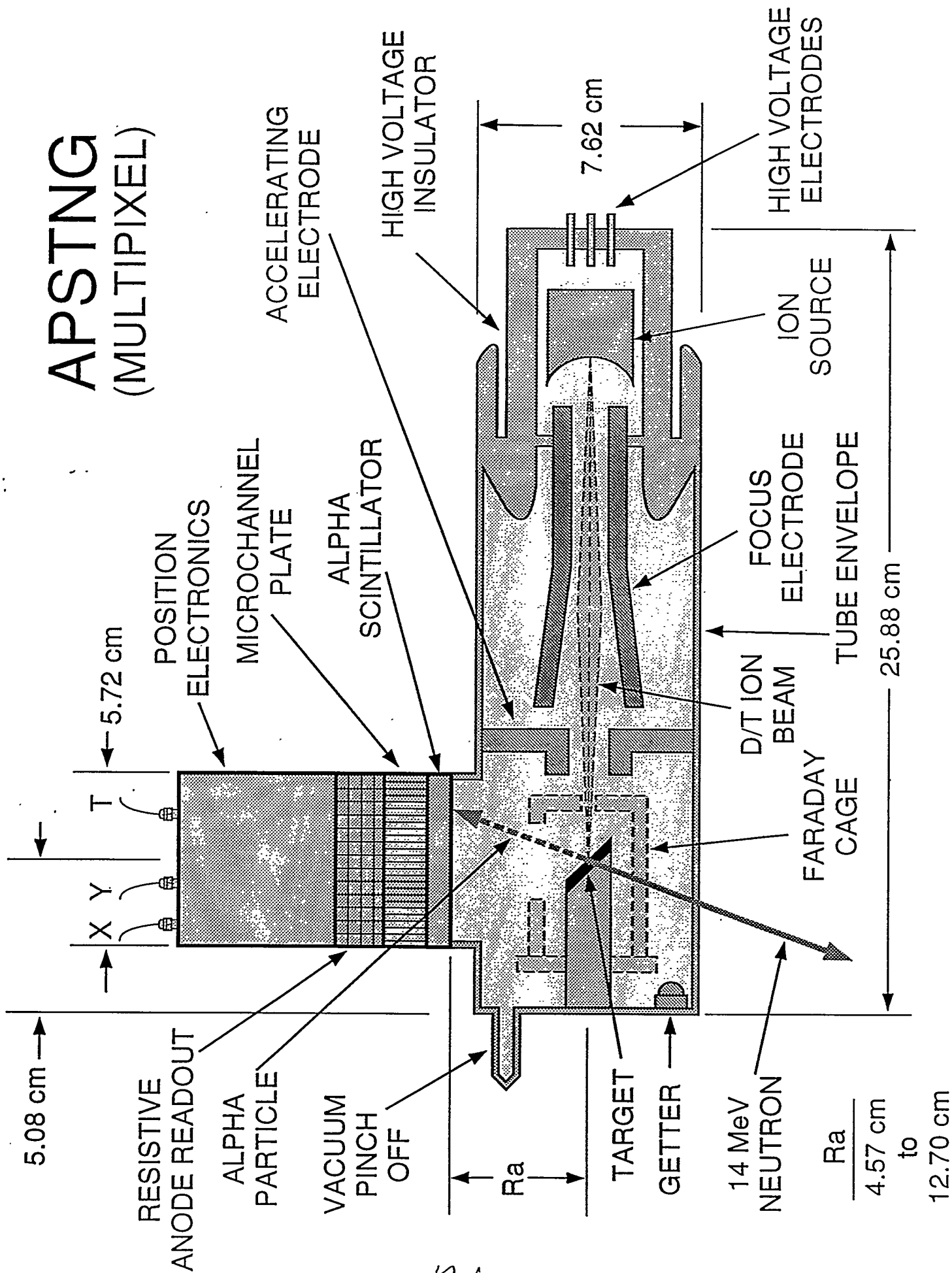


Fig. 2

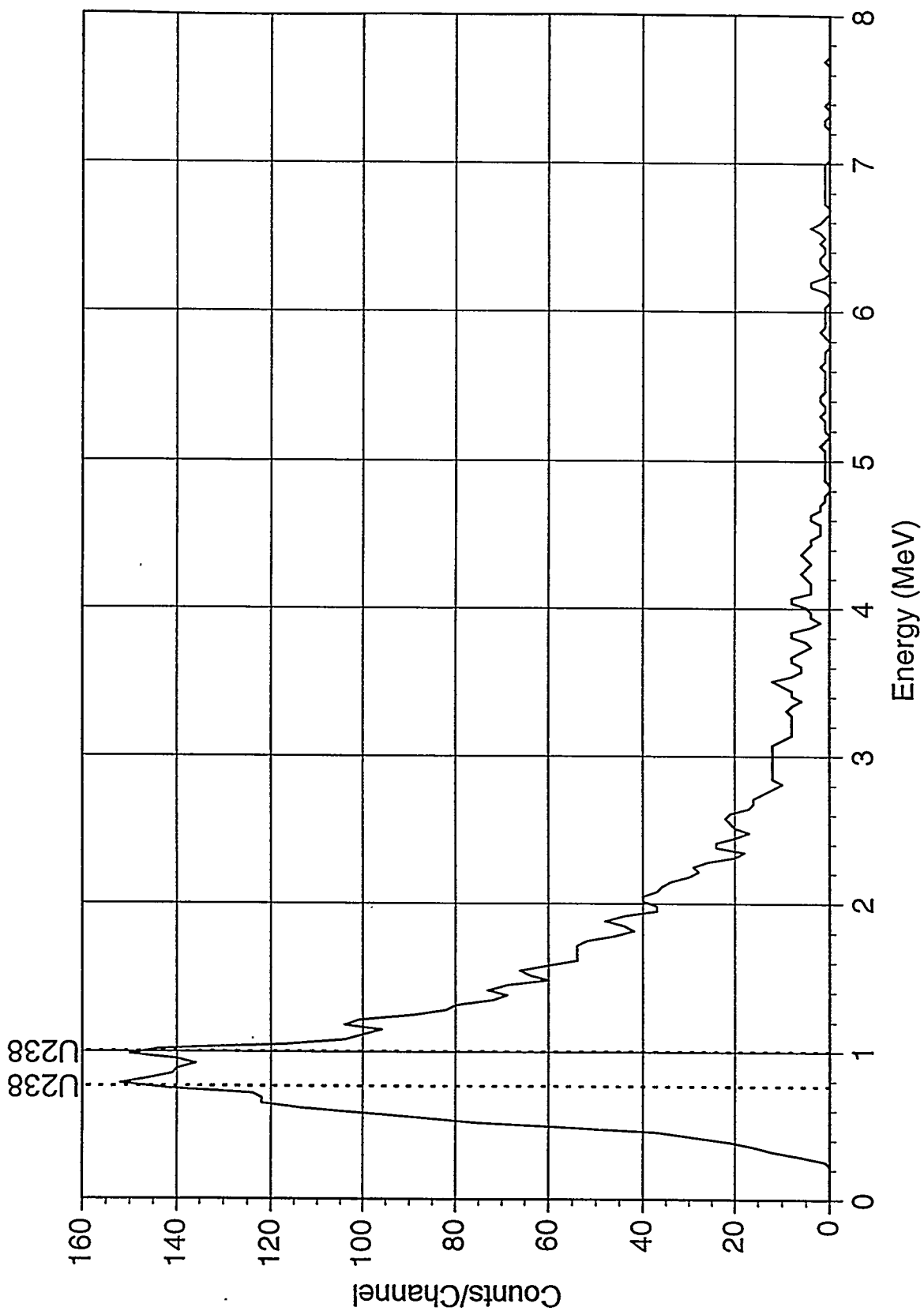


Fig 3

# Chlorine & Depleted Uranium in Concrete (ANL-Drum)

(flight-time channels 60-93)

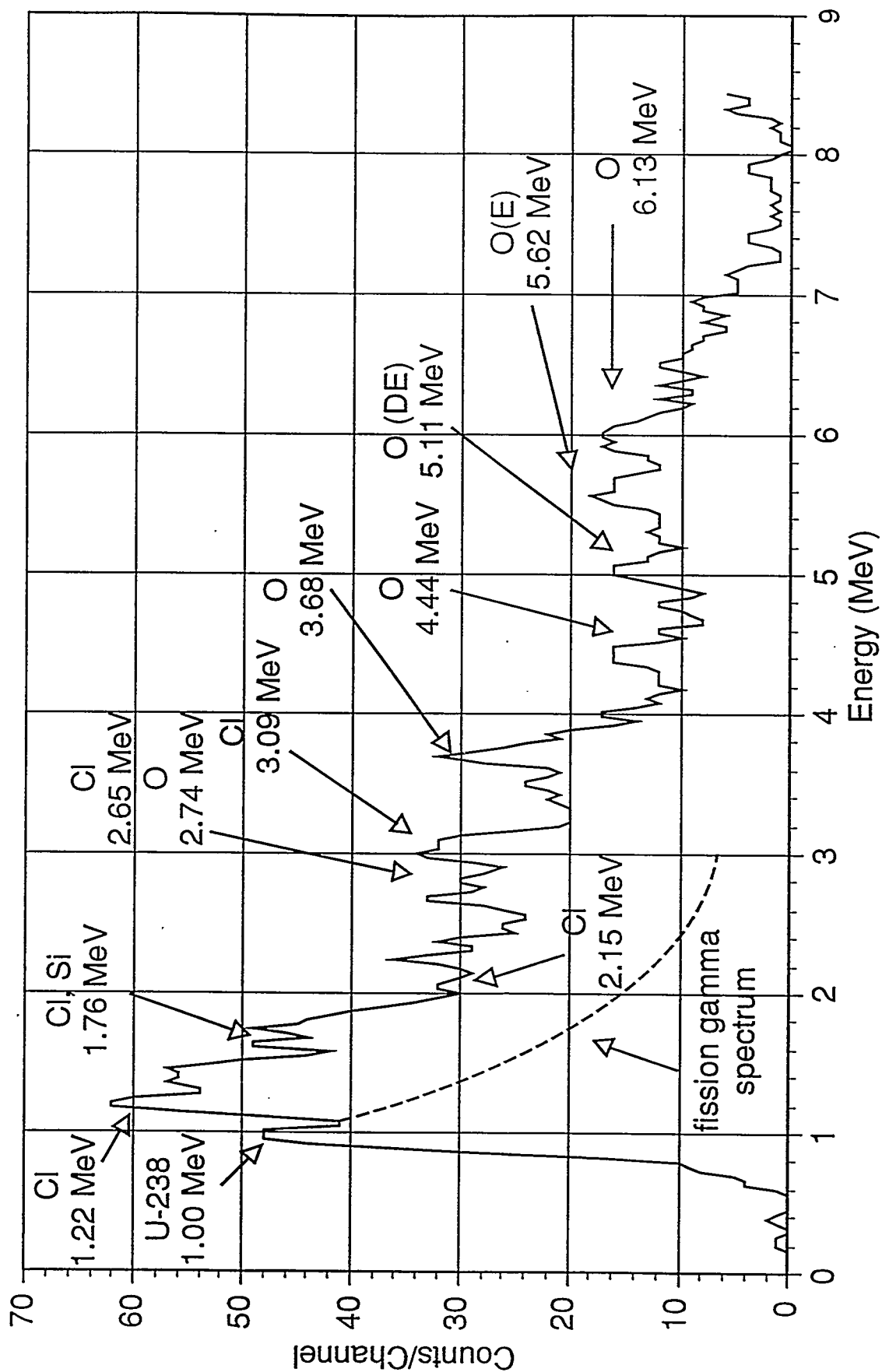


Fig. 4

# Chlorine & Depleted Uranium in Concrete (ANL Drum) (flight-time channels 93-119)

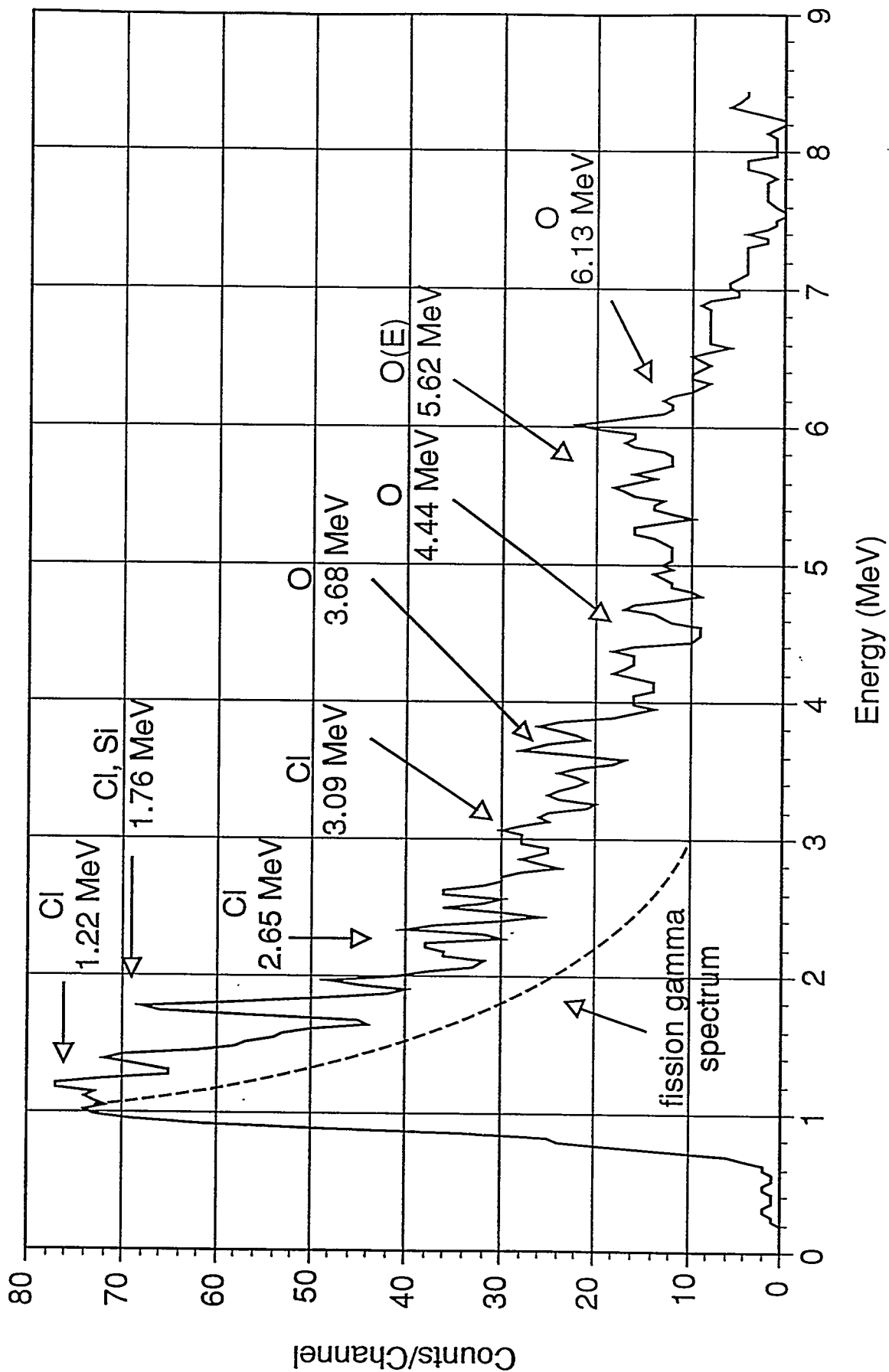


Fig. 5

**PGRS Spectrum of Radwaste Sludge Drum with Pu-239**

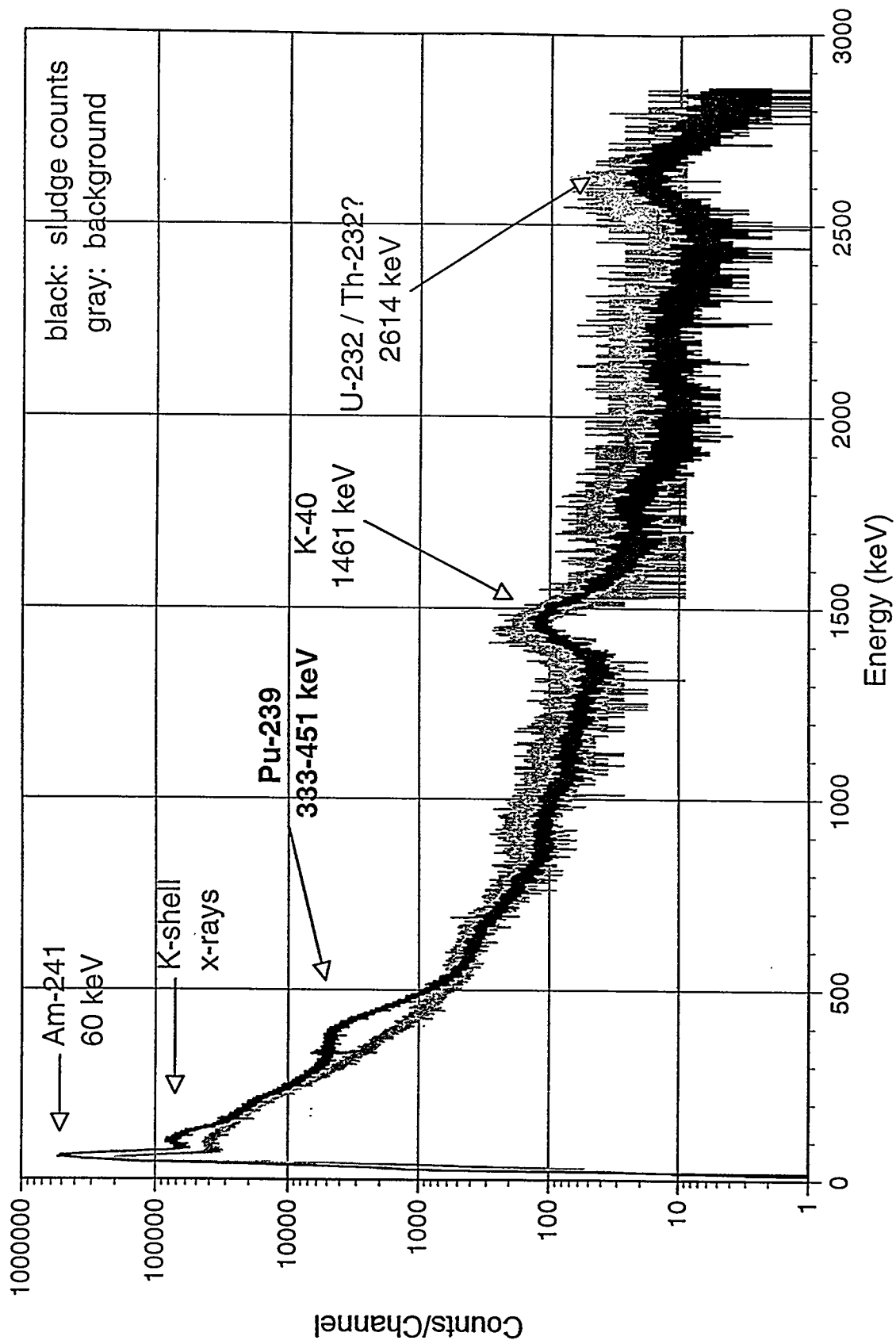


Fig. 6

# PGRS Spectrum of Radwaste Sludge Drum with U-238

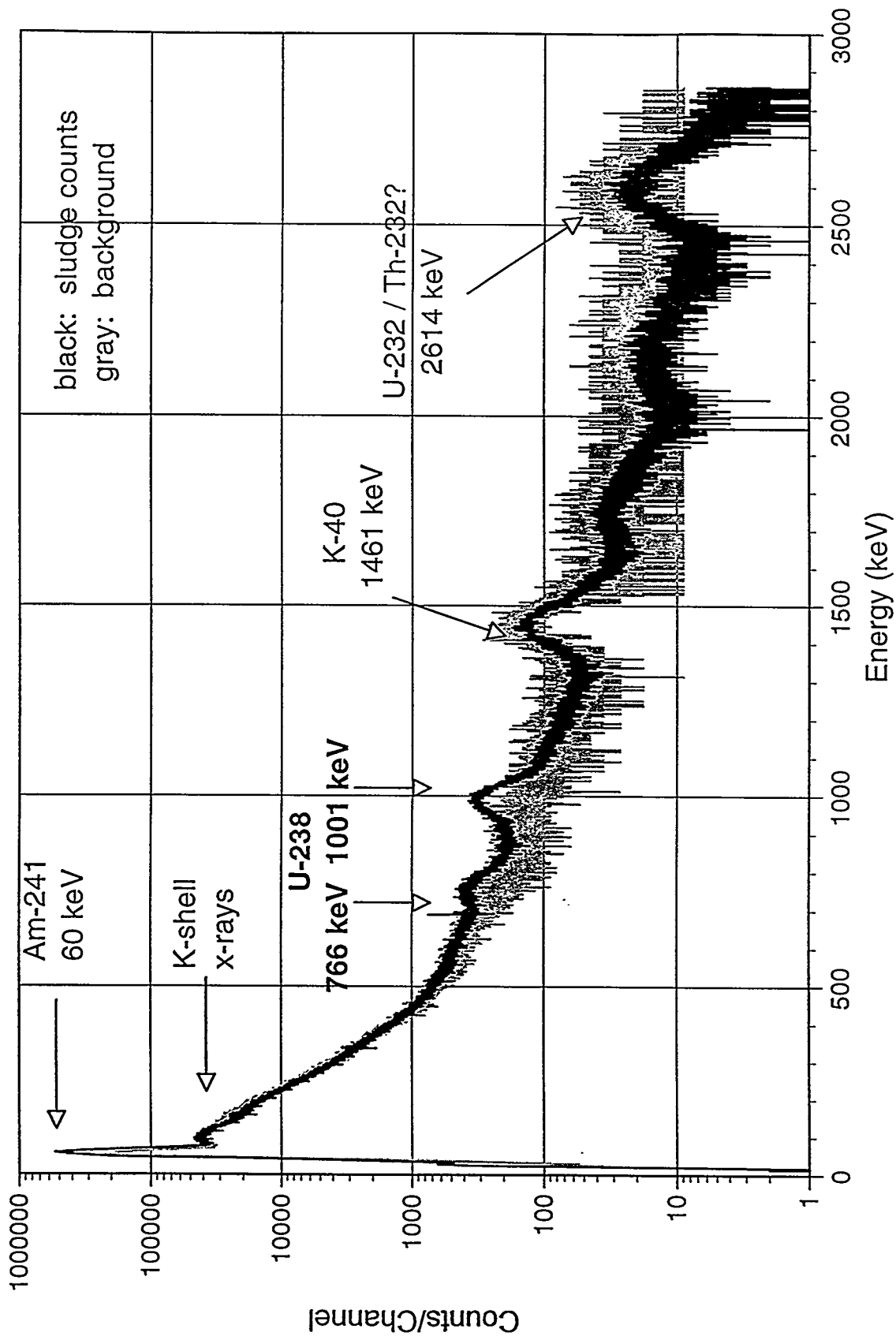


Fig. 7

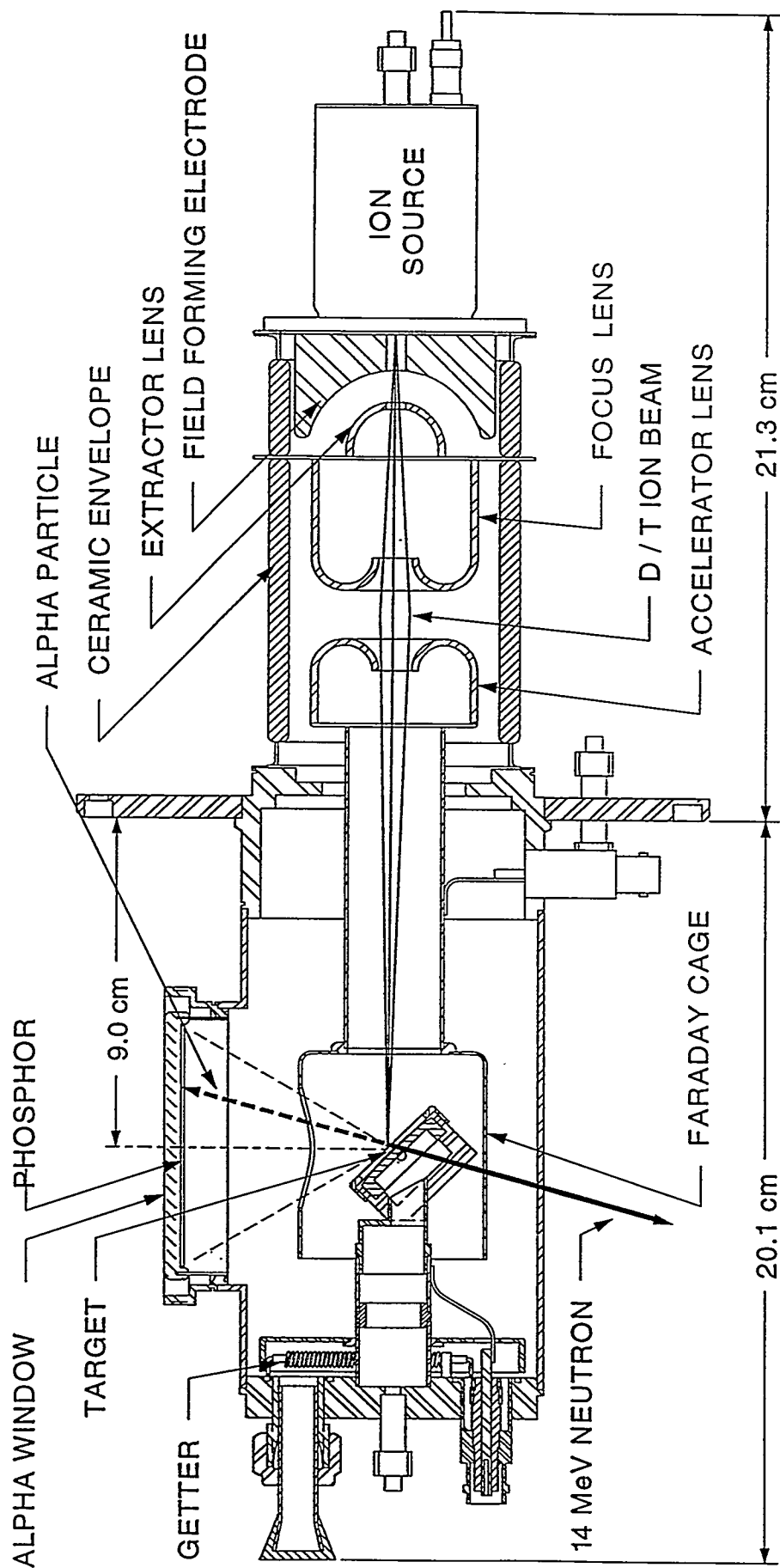


Fig. 8



# MF PHYSICS ACCELERATOR HEAD

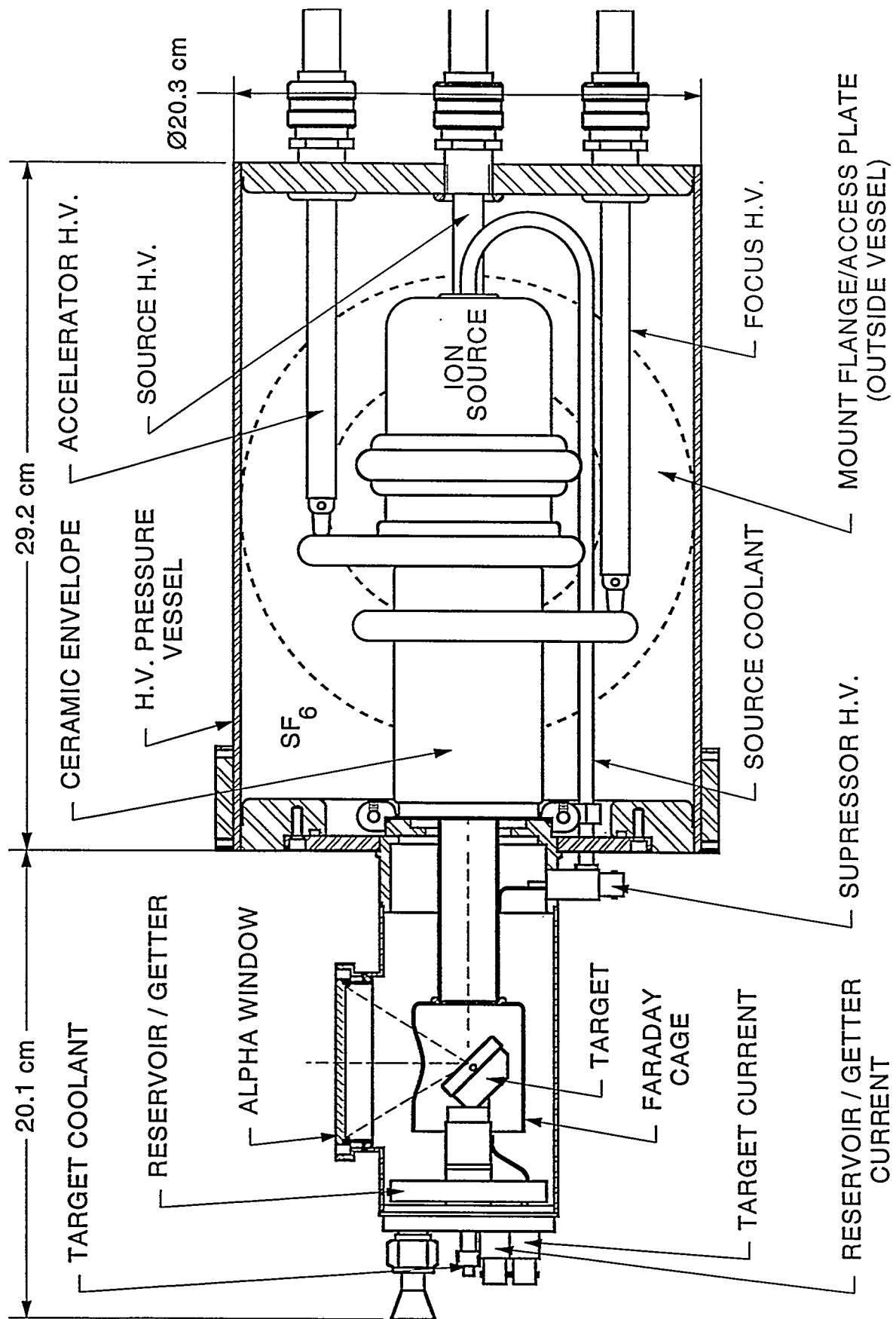


Fig. 9