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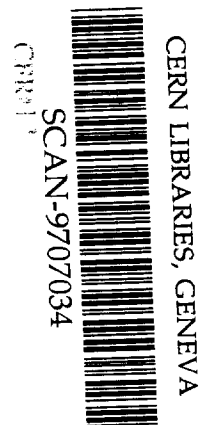
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**ERNEST ORLANDO LAWRENCE
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**A New Intense Neutron Generator
and High-Resolution Detector
for Well Logging Applications**

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P.N. Luke, L.T. Perkins, P.T. Zawislanski,
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A New Intense Neutron Generator and High-Resolution Detector for Well Logging Applications¹

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Abstract

Advances in both ion source and gamma-ray detector technology at LBNL are being used to develop a new high-sensitivity neutron logging instrument. Up to 37 mA of current per 10 - 20 μ s pulse, 80-95% D⁺, has been produced by a 2 inch diameter pulsed multicusp ion source. A D-T neutron flux of 10⁹ - 10¹⁰ n/s is projected from this data. CdZnTe is being developed as a possible gamma-ray detector because of its potential for good energy resolution and efficiency, and ability to operate at room temperature. 3-D time-dependent Monte Carlo calculations show the utility of this system for locating contaminants, especially chlorine-containing solvents, at remediation sites.

I. INTRODUCTION

Neutron logging instruments have been used commercially for decades to locate oil, water, and minerals. In these systems, neutrons produced by isotopic or accelerator sources penetrate the ground, and produce, by neutron capture or inelastic scattering, gamma rays whose energies are characteristic of the elements of the medium.

Recent developments at LBNL in ion source technology and solid state gamma-ray detectors can greatly enhance these systems. By replacing the Penning ion source in conventional D-T or D-D neutron tubes with an rf-driven multicusp source, the neutron flux can be increased by one to two orders of magnitude, to 10⁹ - 10¹⁰ D-T n/s. This flux implies lower detection thresholds, reduced counting times, and holds the promise of significant improvement in spatial resolution through 3-D imaging. Tube lifetime is also expected to improve.

Commercial neutron logging systems typically use either NaI or Ge gamma-ray detectors. While Ge detectors offer the energy resolution (small fraction of a percent) necessary for some applications, they must be liquid nitrogen-cooled, making field applications inconvenient and expensive. NaI offers only ~5% FWHM energy resolution ($\Delta E/E$ at 1 MeV), and though the NaI crystals themselves are relatively rugged, the photomultiplier tubes used for readout are bulky and fragile. At LBNL, innovation in charge sensing electrode design have made possible the use of CdZnTe, a high-Z, wide bandgap semiconductor, for gamma-ray detection. This material offers promise as a rugged, relatively high efficiency, room temperature gamma-ray detector with energy resolution ~1% FWHM.

Neutron source design and performance are discussed in Section II below. Section III contains a description of the status and experimental results of the CdZnTe detector.

In Section IV 3-D Monte Carlo modeling results are presented. These establish lower bounds of detection for chlorine-containing contaminants in the earth, as well as for water in fractured media. The high cross section of chlorine for thermal neutrons, and the small amount of chlorine usually found in uncontaminated media make neutron logging a highly sensitive, quantitative detection method for chlorinated free-phase DNAPL.

II. MULTICUSP ION SOURCE RESULTS

The LBNL multicusp ion source for this application [1] consists of a cylindrical volume plasma source 3.5 cm in diameter and 8 cm long, surrounded by samarium-cobalt permanent magnets (36 mm x 3.5 mm x 3.5 mm), which provide plasma confinement. A porcelain-coated helical rf antenna 1.5 cm in diameter made from 1.6 mm diameter copper wire is used to ionize the plasma. The plasma chamber is terminated by an extraction electrode which in the final accelerator configuration will be followed by an electron suppression electrode. The beam is extracted through a 2 mm diameter aperture in the two electrodes. Because of the suppression electrode, ion current measurements will be accurate and unambiguous.

The rf power supply for the ion source will be located above ground, and the power fed downhole through a 50 Ω coaxial cable. Use of the source is restricted to depths less than about 500 feet unless rf power requirements can be significantly reduced, since a downhole rf power source of 2 to 4 inch diameter is presently beyond the state of the art. An impedance-matching network, consisting of a step-down isolation transformer and an LC tank circuit located adjacent to the ion source, is used to match the impedance from the rf amplifier to the antenna. It is the capacitors in this unit that presently set the minimum diameter of the neutron logging instrument at 2 inches, but this can be reduced using custom-made capacitors.

Ion pulse lengths for the multicusp source are presently power-supply-limited and range from approximately 10 to 30 μ s. Rise time is of the order of 5 μ s, and fall time ~10 μ s. At present the source is run at 100 Hz, due to limitations of the rf power supply. At this repetition rate active cooling is unnecessary. It is not anticipated that the ion source will limit the neutron tube repetition rate, though cooling of the matching network may be necessary at higher pulse rates. One of the chief challenges in reducing the size of the source to downhole dimensions was to lower the operating gas pressure, thus ensuring freedom from electrical breakdown in the accelerator. The source now runs successfully at pressures \leq 5 mTorr. All testing is being done with hydrogen; no significant change is expected for deuterium.

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Measured current density as a function of rf power input is shown in Fig. 1.

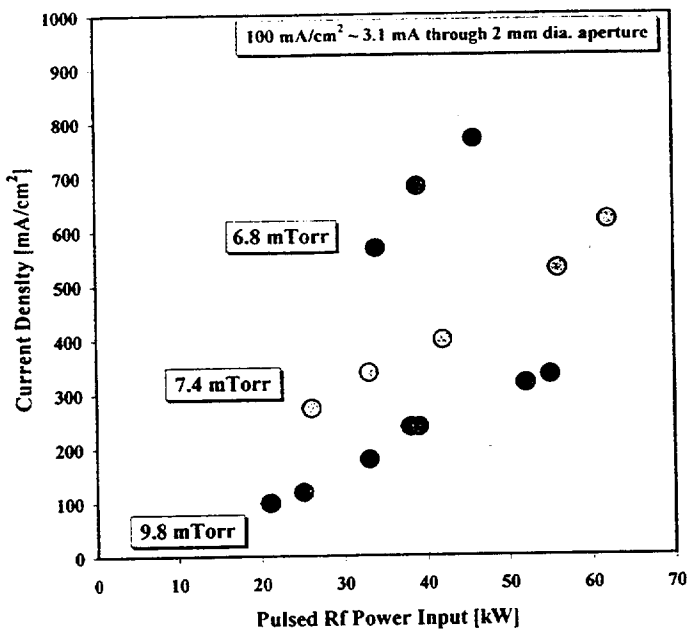


Figure 1. Peak extractable current density at three different source pressures versus rf input power.

Note that for the 2 mm aperture of the tests, 100 mA/cm² gives 3.14 mA (per pulse). Fig. 2 gives the percentage of monatomic, di, and triatomic species in the beam vs. rf power, as measured by a magnetic mass spectrometer.

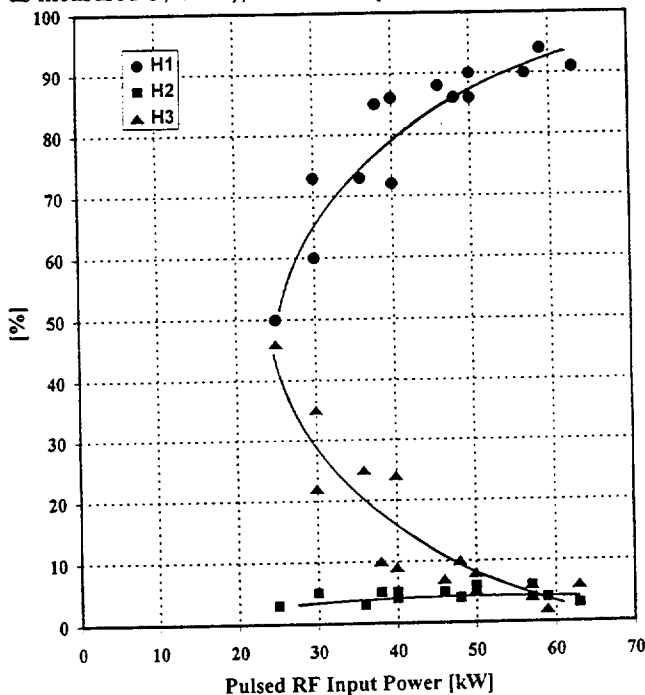


Figure 2. Hydrogen ion species distribution versus pulse rf input power for pressures in the 6 - 9 mTorr range.

Note the high percentage of monatomic hydrogen for higher rf power, giving ultimately a much higher percentage at

the optimal energy for the neutron cross section than in commercial tubes. This percentage can be further increased, though at the expense of current, by use of a magnetic filter.

From the current and species data shown, neutron yield can be projected. Comparing the value of these parameters to those for commercial sources, which produce 10⁸ n/s, we project a yield of 10⁹ - 10¹⁰ n/s, depending on input power, for our prototype source. Higher neutron flux can easily be obtained if the exit aperture is enlarged, since current density remains constant. If the D-D reaction were used, as is desirable for some applications, the estimated yield would be 10⁷ - 10⁸ n/s.

Measurements subsequent to those of Fig. 1 have shown that higher current density as well as monatomic fraction can be attained by optimizing the position of the rf antenna. Other possible improvements include the addition of a discharge starter, to reduce the input rf power and source operating pressure.

The high neutron yield just described is desirable for many applications, e.g., detection of unexploded mines. For some downhole applications, however, the resulting gamma flux may exceed the count rate limit of the detector. For these applications the source would be run at much higher pulse rates (~1-10 kHz) with the number of neutrons per pulse is reduced to acceptable levels (~10⁵ - 10⁶ n/pulse). The average flux produced (10⁹ n/s) is then approximately an order of magnitude higher than that currently attainable in commercial systems. Reducing the instantaneous ion current is usually accomplished by reducing the input rf power. However the reduction desirable here is so great that ionization of the plasma would not proceed. Therefore the current reduction would be accomplished by reducing the accelerator voltage.

At this time a 100 kV accelerating system is being designed for the neutron source. The IGUN[®] code has been used to calculate an ion optics solution which appears to spread the beam uniformly on the target. This is expected to extend target and overall tube lifetime by reducing target heating and sputtering, which leads to electrical breakdown.

III. GAMMA-RAY DETECTOR DEVELOPMENT

For this project, we have chosen to develop a new type of semiconductor detector for the detection of neutron induced gamma rays. This detector will offer the advantages of significantly better energy resolution than NaI and, unlike Ge, operation at room temperature. For over 20 years, a great amount of effort has been spent in the development of room temperature detectors based on the use of wide band-gap, high atomic number semiconductors. Much of the attention has been concentrated on the materials HgI₂ and CdTe. Despite significant progress, several important problems remained. Foremost was the generally poor charge transport properties of these compound semiconductors. In particular, the transport of holes is much worse than that of electrons. This results in signals that vary in amplitude depending on the location of gamma ray interaction, thus adversely affecting the detectors' energy resolution. This factor, combined with limitations on crystal sizes, the presence of polarization effects, and material

processing difficulties, prevented these detectors from being widely used.

Recently, a new semiconductor material, $\text{Cd}_{(1-x)}\text{Zn}_x\text{Te}$, has been developed for detector applications. It is grown using the high-pressure Bridgman process, with Zn concentration (x) ~ 0.1 . This material shows substantial improvement over previous semiconductors - large crystals can be grown, and detectors fabricated from this material do not exhibit significant polarization effects. Charge transport properties, however, remain comparable to those of CdTe and thus the energy resolution obtained using conventional detector configurations is still poor. In order to address this problem, our detector group has developed a new charge sensing technique. This technique uses a coplanar grid electrode configuration on the detector, together with signal subtraction, to preferentially sense the collection of electrons [3]. This minimizes the effects of poor hole transport and, additionally, compensation can be made for electron trapping. Using this technique, dramatic improvements in energy resolution have been demonstrated. An energy resolution of 2.4% FWHM for 662 keV gamma rays has been achieved for a 1 cm^3 coplanar-grid CdZnTe detector. Much better energy resolution, to $<1\%$ FWHM, is in principle obtainable with this method [4].

Currently, the maximum practical detector volume is $\sim 2 \text{ cm}^3$, a limit determined by the size of CdZnTe crystals that are available with sufficiently uniform characteristics to achieve the desired detector performance. On the other hand, because of the higher average atomic number of CdZnTe compared to Ge and NaI, the detection efficiency of a CdZnTe detector is significantly higher than that of a Ge or NaI detector of comparable volume. The calculated photopeak efficiencies of CdZnTe, Ge and NaI for varying detector volume are shown in Fig. 3.

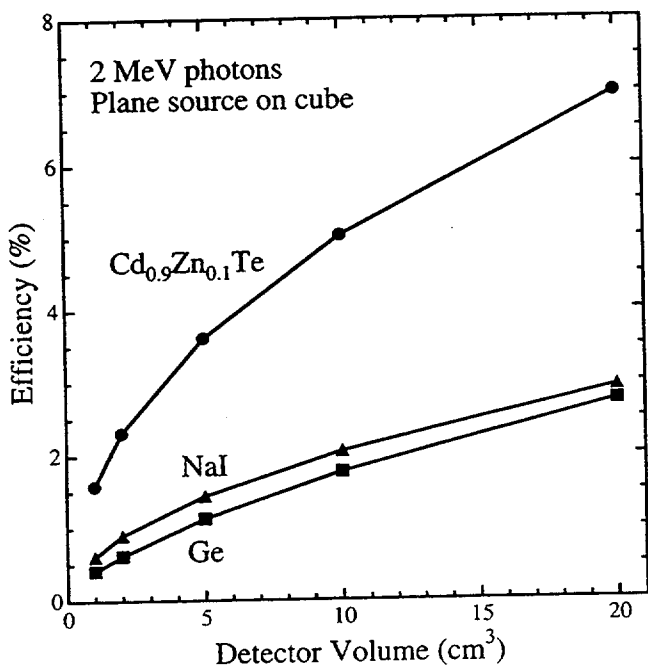


Figure 3. Calculated photopeak efficiencies of CdZnTe, Ge and NaI as a function of detector volume for 2 MeV photons.

These were calculated using the pulse height tally option of the code MCNP [5] (see section IV), where the energy deposited by each source particle and its secondaries in the detector is recorded and normalized to the total number of source particles. The results for Figure 3 assume a plane of 2 MeV photons incident on one face of the detector cube. All source particles enter this face. The efficiency is determined by taking the total counts in the full energy photopeak (2 MeV in this plot) and normalizing to the total number of source particles sampled by the Monte Carlo calculation. These results indicate that for an equal detector volume, CdZnTe is about three times more efficient at 2 MeV than either Ge or NaI. Efficiency calculations for higher energy photons of importance for capture gamma ray detection indicate similar relative efficiencies for these detectors.

The goal of the present work is to develop coplanar-grid CdZnTe detectors with volumes $\sim 2 \text{ cm}^3$ and energy resolution of $\sim 1\%$ FWHM. This will provide a factor of ~ 5 improvement in energy resolution compared to scintillators. Modeling results show that CdZnTe detectors with such volumes will be already count-rate limited due to the high neutron flux obtainable from our new neutron source. Higher count-rate throughput, and thus higher detection efficiency, may be achieved using multiple detectors.

One concern regarding the use of CdZnTe detectors in neutron logging applications is the effect of the neutrons on detector performance. Cd has a very high cross section for thermal neutron capture, which could give rise to unacceptably high background levels. As discussed in Section IV, our calculations have shown that neutron shielding can be used to effectively diminish thermal neutron interactions in the detectors and an acceptable background level can be achieved.

Another issue is the effects of radiation damage. Fast neutrons can cause damage to the crystal lattice of the detector, thereby degrading the charge collection properties and adversely affecting its energy resolution. Radiation damage effects in conventional CdZnTe detectors have been observed in a limited number of experiments. The threshold fluence for observable damage from intermediate energy neutrons from a ^{252}Cf source has been found to be $\sim 10^{10} \text{ n/cm}^2$ [6]. For our application, such a damage threshold would seem to be acceptable. Nevertheless, radiation damage experiments using coplanar-grid CdZnTe detectors will need to be conducted to realistically assess the suitability of such detectors for neutron logging applications.

A detector module that contains the detector and front-end electronics has been fabricated. The module is air and water tight to facilitate operation in the field and in borehole environments. The power consumption of the front-end electronics is less than 150 mW, which minimizes self-heating and allows the detector module to be battery powered. The output of the detector module interfaces directly with a portable amplifier/multi-channel analyzer for data acquisition. Fig. 4 shows the spectral performance of a 2.25 cm^3 CdZnTe coplanar-grid detector that is being developed for use in this system.

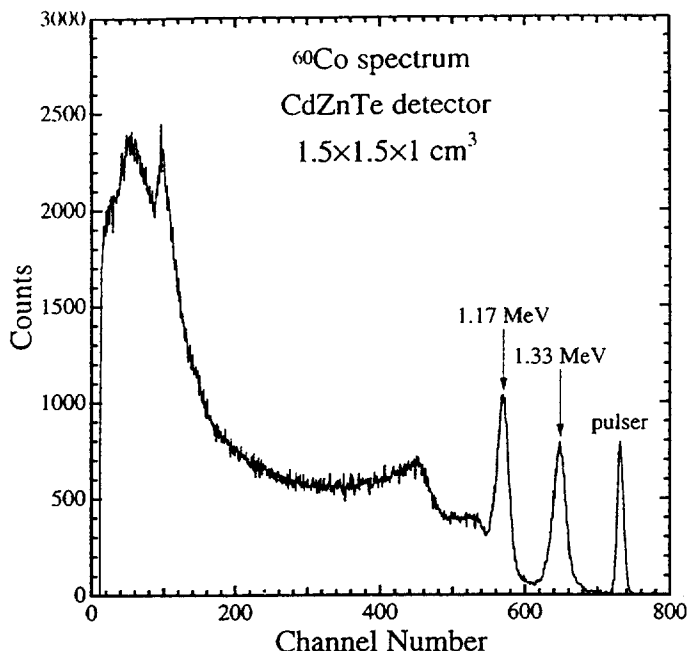


Figure 4. ^{60}Co spectrum obtained by a coplanar-grid CdZnTe detector.

The energy resolution at 1.33 MeV is about 3.1% FWHM, which is slightly better than NaI detectors. We believe the resolution of this detector is limited by spatial non-uniformity in the charge transport properties of the material. Significantly better resolution should be obtainable with better materials and further optimization of the coplanar-grid technique.

IV. MODELING AND APPLICATIONS

Though we believe the logging system described here to be useful for a number of applications, including detection of environmental pollutants in the earth (chlorine and cadmium, for instance, are known to have large neutron cross-sections), unexploded mines (D-D neutrons), petroleum, and water, we have as yet only explored briefly the first and last of this list. For this study the 3-D point-energy, time-dependent, Monte Carlo code MCNP, with the ENDL60 [7] cross section library, was used. Both water and DNAPLs can be detected using the gamma rays promptly emitted by H and Cl when these elements absorb low energy neutrons. As is characteristically done in neutron logging, the detector electronics must be gated off for the first few tens of microseconds of the pulse in order to reduce the gamma ray background and to eliminate count-rate limitation problems due to inelastic scattering gamma-rays. Following a parametric study of the effect of the gate width, a value of 30 μs was chosen for the runs described below.

The example used for DNAPL detection was the detection of TCA-- $\text{CHCl}_2\text{CH}_2\text{Cl}$. Two cases were considered: DNAPL uniformly distributed in a medium of wet sand (70% silica and 30% water by volume), and DNAPL in fractured granite. For the first case, the composition of the sand was as follows (% by weight): SiO_2 (72%), Al_2O_3 (15%), Fe_2O_3 (2%), FeO (3%), MgO (1%), CaO (1%), Na_2O (3%), K_2O (2%) and CO_2 (1%). Its specific density (for 30% porosity) was 1.9 g/cm^3 .

Fig. 5 shows the flux spectrum of 7.3 to 7.6 MeV photons at the detector volume for different TCA concentrations in sand.

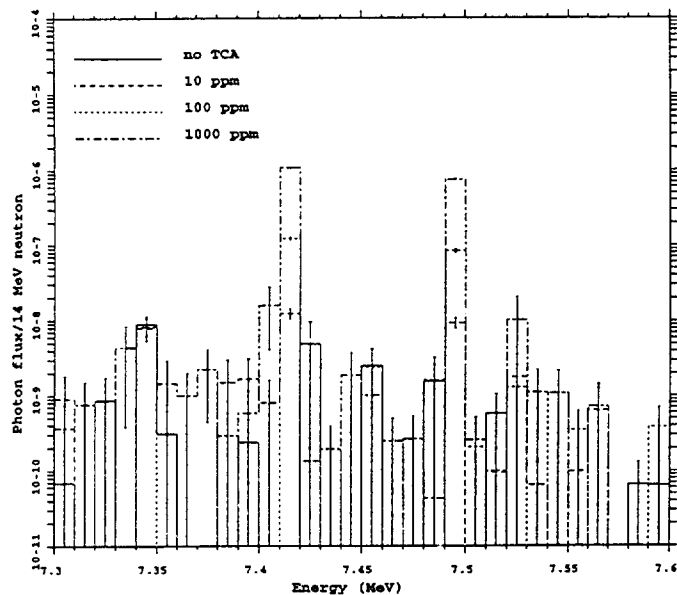


Figure 5. Effect of TCA concentration in water on the photon spectrum in the detector volume. Medium is made of sand (1.9 g/cc) and water (0.3 g/cc).

For these calculations the homogeneous medium was penetrated by a 10 cm diameter borehole, at the center of which was a 2 cm radius spherical CdZnTe detector, surrounded by a 0.5 cm neutron shield of $^{10}\text{B}_4\text{C}$. A D-T point neutron source was located 4 cm above the detector center. Examining the 7.414 and 7.495 MeV Cl lines it is seen that even a concentration of 10 ppm TCA in water should be detectable with a good resolution and good efficiency detector. A similar conclusion can be drawn from the 6.111 MeV peak of the Cl spectrum.

For the detection of TCA in fractured granite the same model was used for the detector, source, and borehole. Vertical fractures with a range of fracture widths and distances from the borehole center were studied. For granite containing 0.1 g/cm^3 water, the detectable range for 1 cm fractures filled with TCA was about 40 cm, and for 1 mm it was approximately 30 cm.

Detection of water in fractured granite was studied using the same model described above for TCA. Measuring the 2.23 MeV hydrogen peak, it was found that 1 cm fractures at a distance of 20 cm could be detected in dry granite, but the range was only 10 cm with 0.1 g/cm^3 water in the granite. Adding 0.3 g/cm^3 NaCl to the water, which is possible in many field experiments, makes it possible to detect a 1 cm fracture at 40 cm by measuring the 6.1 MeV Cl (n, γ) peak, even if the rock contains 0.1 g/cm^3 fresh water. For 1 mm fractures the corresponding limit is approximately 20 cm.

A small study was done to estimate the effect of the source - detector distance on the signal amplitude. It was found that, relative to the closest approach situation used above, displacing the source by 30 cm along the borehole decreased the signal per neutron by approximately 25%. For 60 cm, the reduction was 50%.

We have begun a study of the localization ability of the system. As a first example, a 2 cm radius spherical CdZnTe detector was encased in two spherical shells—a 2.5 cm thick photon-absorbent layer covering the 0.5 cm thick $^{10}\text{B}_4\text{C}$ neutron shield mentioned above. A 15° pole-to-pole wedge

was cut from the photon shield to form a collimator. Lead and tungsten were explored as materials for the photon shield. It was found that lead gave the better performance-- tungsten produced a large flux of secondary photons. A vertical fracture intersected the center of a borehole in dry granite. The fracture, though not the borehole, was filled with water. Fig. 6 shows a drop in signal of a factor of 3 as the gap is turned 15 degrees from the fracture.

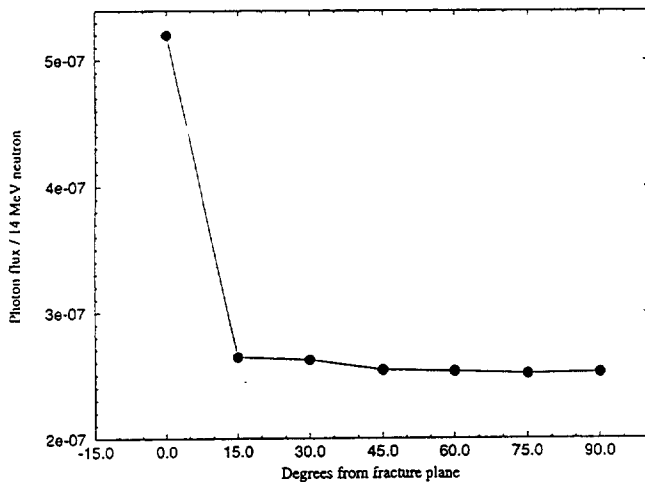


Figure 6. Angular dependence of the 2.23 MeV photon flux in a directional detector, due to water in 1 cm wide vertical fracture intersecting the detector center. Zero degrees corresponds to the fracture plane. The medium is dry granite.

This shows promise for some imaging of chlorine by the system, to be obtained by taking readings as the detector is rotated at each of several different depths.

V. SUMMARY AND CONCLUSIONS

A miniaturized multicusp ion source for neutron logging applications has been fabricated and tested. Due to its high current (up to 37 mA/pulse) and the high monatomic species content (~90%) of the beam, it is projected to yield $10^9 - 10^{10}$ n/s.

A new coplanar electrode system has enabled the development of CdZnTe gamma-ray detectors for this application. These detectors show great promise as relatively high efficiency, rugged, room temperature detectors, with energy resolution expected to be approximately 1% FWHM. 3-D Monte Carlo calculations show that a thin shield of ^6LiF or $^{10}\text{B}^4\text{C}$ can be used to effectively eliminate secondary photon production due to neutron capture in the detector. The code MCNP has been used to study application of the neutron logging system to the detection of DNAPL and water. Results show that quantitative measurements of free-phase DNAPL can be made, with sensitivity of ~10 ppm expected in

sediment containing 0.3 g/cm^3 water. DNAPL-filled fractures of 1 mm width can be detected at a distance of about 30 cm from the borehole.

For detecting water, a much more sensitive measurement can be made if the water is saturated with NaCl, so that the Cl peak can be detected. In this case mm-sized water-filled fractures in granite with 0.1 g/cm^3 structural water can be seen at a distance ~ 20 cm.

Localization of detected substances such as DNAPL looks very promising, based on preliminary studies of system angular resolution. However much more modeling must be done, followed, of course, by laboratory experimental studies, in order to determine the effect of variable lithology on the results.

VI. ACKNOWLEDGMENTS

The authors gratefully acknowledge the contributions of N. Madden and C. Cork, who designed and built the electronics and housing for the CdZnTe detector module.

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