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DISCLAIMER

Limits on Electromagnetic and Particle Emission from Palladium-D_2O Electrolytic Cells

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We have carried out a series of experiments to test the recently claimed observations of cold fusion. In a first experiment we used an electrochemical cell with a Pd wire cathode. Neutrons were detected by the $^1H(n,\gamma)$ photons from surrounding water and paraffin. In a second experiment, two vacuum-cast Pd disks were the cathodes in "twin" cells, one with H_2O, and the other with D_2O. The two cells were shuttled every 24 hours between similar detector setups, equipped with neutron, γ- and X-ray detectors. A third experiment was designed to measure production of charged particles, using a Si surface barrier detector viewing the back of a 76 μm thick Pd foil cathode. No statistically significant evidence of nuclear fusion has been obtained in any of these experiments, either in steady state operation or in transient response to a variety of perturbations. Upper bounds on the rate of various postulated fusion processes were set including $< 2 \times 10^{-23}$ neutrons (dd pair)$^{-1}$ s^{-1} for the d(d,n)^{3}He reaction and $< 6 \times 10^{-25}$ protons (dd pair)$^{-1}$ s^{-1} for the d(d,p)^{3}H reaction (2σ level). Chemical analysis of our electrolytes revealed no anomalous increase in ^3H concentrations.

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I. INTRODUCTION

The recently reported claims of possible observation of cold fusion[1,2] of deuterium in Pd and Ti cathodes have generated much interest. Shortly after these initial announcements were made, we initiated a series of experiments to test for "cold" fusion in similar electrochemical cells. We have chosen to follow the Fleischmann and Pons (FP) approach[1] since they reported the largest energy, neutron and tritium production rates. Initially we carried out a simple experiment with a cell constructed with a thin Pd wire cathode. Following this we designed a more thorough "blind" experiment equipped with a host of radiation detection systems for examining fusion reactions involving the emission of neutrons and/or radiations from fusion reactions not normally considered (e.g. high energy e^+e^- pair and conversion electrons). In addition, we have designed a special cylindrical cell with a thin Pd cathode window to search for production of protons from the d(d,p)t fusion reaction channel. A similar type of experiment is described in a recent publication by Ziegler et al[3].

The cells in these experiments were all operated as open systems. In the following sections we describe the design, preparation, treatment and operation of the electrochemical cells, followed by presentation and discussion of the results from these experiments.

II. Pd-WIRE EXPERIMENT

A. Cell design and electrochemistry

In this experiment the electrochemical cell was of a simple, undivided design. A three electrode configuration was employed consisting of:

- a 0.25 mm dia. platinum wire electrode (anode), coiled in a 17 mm dia., 11 turn spiral of 5 mm pitch, coaxial with the working electrode.
- a palladium wire working electrode (cathode), 0.25 mm dia., 302 cm length, wrapped in a tight coil about a soft glass melting point capillary, 3 layers with a total mass of 1.9440 g. In addition, 0.7576 g of Pd "black" was plated *in situ* from a tetracyanopalladium (IV) solution onto palladium, making the total mass of the working electrode 2.7016 g (0.02539 moles).
- a palladium reference electrode, poised at α/β-PdD_x.

The electrolyte was 1.0 M LiOD in D_2O (Cambridge Isotopes Laboratories, lot # F3079, 99.8% D, and BioRad Laboratories, lot # M1152, 99.8% D), made by reacting elemental Li with heavy water. Total cell volume was 12 mL. Due to electrolysis and evaporative losses, frequent "topping up" of the electrolyte using

pure D_2O was required. The rate of water loss was about 2–3 times that calculated based on electrolysis only.

At the start of the experiment, the electrode surface was cleaned of adsorbed material and activated using a square wave current cycle (±500 mA, 10 Hz) for 90 minutes. Following this treatment, the electrode was saturated with atomic deuterium by controlled current electrolysis. A regimen of steadily increasing applied current was followed, starting with 1 mA, increasing in a 1,2,3,5,10... sequence, and ending with 1.25 A approximately 18 hours later. Thereafter, current was maintained at a level between 250 mA and 1.25 A; commonly i=500 mA was used overnight to avoid excessive electrolyte loss while the cell was unattended, with the current being increased during the day to its upper limit.

After the palladium was saturated with deuterium, several surface poisons were tested for their effectiveness in decreasing the rate of deuterium atom recombination at the Pd surface, thereby increasing the overpotential at a fixed current density. Lead (II) in the form of basic lead acetate was found to be effective at the monolayer level, as was thiophene (25μL in 12 mL electrolyte).

The open-circuit potential was monitored at regular intervals, as was the working electrode potential during electrolysis. The high values of overpotential observed during the electrolysis were found to be largely kinetic in origin; the open-circuit potential did gradually move negative of the α/β Pd deuteride equilibrium potential, but not by more than a few hundred millivolts. The implication is that an inordinately high degree of supersaturation of deuterium in the palladium was not taking place as has been suggested in some accounts[1]. This finding was supported by the electrochemical titration of absorbed deuterium performed at the end of the experiment.

After 24 days of electrolysis, the PdD_x electrode was purged of deuterium by controlled-potential electrolysis *in situ*. Consistent with surface deactivation by adsorbed thiophene, a significant overpotential (1.0V) was needed to remove the deuterium from the metal. There was some small loss of deuterium as D_2 gas in the first few minutes of electrolysis, so that the estimate of absorbed deuterium is only a lower bound. A total charge of 1518 Coulombs was passed, leading to an equivalent mean stoichiometry of $PdD_{>0.62}$ for our sample. The post mortem analysis of the cell revealed that all Pd wire and Pd black in the electrolyte had been structurally changed (the material became dull grey and brittle), implying that deuterium had penetrated all of the Pd wire. In a control experiment performed after the electrode was purged of deuterium electrolytically, the cell was run under identical conditions in reverse, at the same applied currents, "topping up" of heavy water, etc. This permitted collection of background count rates for the detection

systems under identical conditions to those during the actual experiment.

B. Radiation detection

To detect neutrons we relied primarily on detecting the 2224-keV γ-ray from the ^{1}H(n,γ) reaction. A 20% intrinsic Ge detector system was used for this purpose. It was placed inside an approximately cubical enclosure ($\approx$ 30 cm length), with the cell placed in the middle of the enclosure some 10 cm away from the face of the detector. To enhance the thermalization of neutrons, and thus the yield of the 2224-keV γ-ray, we filled the enclosure with bottles of distilled water and paraffin wax blocks. The enclosure was surrounded by a 5 cm thick layer of lead to lower the γ-ray background in the detector. The main background component that limited the sensitivity for detecting the 2224-keV γ-ray line was the Compton edge of the 2614.5-keV γ-ray from environmental ^{208}Tl. To reduce the ^{1}H(n,γ) background from cosmic-ray induced neutrons, we covered the top of the enclosure with Cd sheets on top of which was placed a 5 cm thick layer of polyethylene bricks.

The efficiency of the Ge detector for detection of neutrons was estimated by placing a ^{252}Cf source (2000 sf/min) at the location of the cell. This calibration indicated an efficiency of $\sim 1.5 \times 10^{-4}$ for fission neutrons. Energy calibration of photons before and after the actual experiment was obtained with standard gamma sources (^{60}Co, ^{137}Cs etc.) in addition to internal calibration from known environmental background γ-rays (511-keV annihilation radiation, 1461-keV from ^{40}K, 2615-keV from ^{208}Tl, etc). Gain shifts throughout the counting period were less than $\pm$ 1 channel ($\sim$ 2 keV). Data were recorded every $\sim$ 12 hours, and gain-shift compensation was carried out during off-line analysis.

A plastic scintillation particle detector placed directly in front of the cell, on the side opposite to the Ge detector, served as a monitor for possible high-energy charged particles (primarily e^-e^+). We also used a variety of BF_3 neutron counters to measure the neutrons directly. In our experiment, the BF_3 detectors proved to be unreliable and were abandoned midway through the experiment.

III. "TWIN" Pd CYLINDER EXPERIMENT

A. Design and electrochemistry

For the second series of experiments we chose a design that allowed measurement of a D_2O cell in parallel with a control H_2O cell. The construction and operation of the two cells were identical, with the exception of H/D substitution in the electrolyte solution, thus the label "twin" for the experiment. To ensure against subjective bias in data analysis, the experiment was run in a "single blind" mode, i.e., the two cells

and corresponding electrolyte solutions were labeled A and B, and their identity was kept secret from experimenters until completion of the experiment and primary data analysis.

The Pd electrodes of the cells were made from vacuum melted and cast cylinders 0.98 cm in dia., and 0.545 cm thickness. The masses of the these electrodes were closely matched (4.5022 g, and 4.5112 g, A and B respectively). The electrodes were spot welded to "nooses" of 0.25 mm diameter Pd wire, and mounted in identical cylindrical plastic cells (50 mL centrifuge cones). Nearly identical cylindrical Pt mesh counter electrodes were symmetrically disposed around the Pd cathode. Electrolyte solutions were 1.0 M LiOH/D, prepared as in the first experiment. Electrolysis was run at 50 mA for H/D saturation (3 days), current was then increased to 100 mA for 6 days, to 250 mA for 2 days, then finally to 400 mA for the remainder of the experiment. Total electrolysis time was 572 hours. Sodium sulphide (10 mM) was added to both cells after saturation to reduce H/D recombination at the surface.

B. Radiation detection

To ensure that radiation measurements from the two cells were taken using the same detectors under similar physical conditions, the "twin" cells were shuttled between two detection setups every 24 $\pm$1 hours. Each of the two setups was made of a shielded enclosure, filled with water and paraffin to thermalize neutrons. The cells were located approximately in the middle of the enclosures and were surrounded by various neutron, photon and charged particle detectors.

Fig.1 shows a schematic diagram of the two detection setups and the physical configuration of the cells and the various detectors. For γ-ray detection we used two 20% Ge detectors with an energy range 0.1–4.7 MeV in setup-I and 0.1–5.7 MeV in setup-II. For X-ray measurements we used a 12 mm thick intrinsic Ge detector with a lower threshold of 10 keV. In setup-I we used an NE213 liquid scintillator neutron detector with pulse-shape discrimination (PSD) electronics to suppress γ-ray detection, while in setup-II we used a high-pressure ^{3}He neutron spectrometer[4]. A paddle plastic scintillator placed over the ^{3}He spectrometer was run in scaler mode to monitor cosmic radiation and possible high-energy charged particles. The detectors were calibrated with X-ray, γ-ray and neutron (^{252}Cf) sources situated as close as possible to the normal position of the Pd cathodes.

IV. THIN Pd FOIL CELL

The electrochemical cell was of a simple cylindrical design (see Fig.2). The pal-

ladium cathode was a 76 μm foil sealing the bottom of the cell. It was first loaded and purged of hydrogen and deuterium electrolytically several times before a 100 nm gold H/D diffusion barrier was evaporated on the side facing away from the electrolyte. The other side of the foil was etched to facilitate H/D incorporation and bubble evolution. The total mass of the active portion of the Pd electrode was 0.272 g, or 0.00255 mol. The electrolyte was a 1 M LiOD in D_2O as in the first experiment and electrolysis was performed at constant current (i=100 mA, A = 2.95 cm^2). Below the Pd window, a Si surface barrier (SSB) charged particle detector was placed 4.6 mm behind the Pd electrode. A continuous dry nitrogen stream was used to flush the detector head space.

The experimental protocol called for taking data with the Pd foil acting as a cathode, saturated with deuterium, and to take "background" data with the foil acting as cathode saturated with H. Both heavy water and light water electrolytes were used. Reverse current controls were not run because of dimensional changes in the PdD_x-to-SSB distance due to the Pd foil deformation following H/D incorporation.

V. RESULTS AND DISCUSSION

A. Pd wire cell

Differential and cumulative counting rates for the 2224-keV neutron capture γ-ray together with those for the background peaks were extracted from data collected for about a two week period and compared with rates from background measurements before and after the run. Post-run background counting (control) was done with the cell run under reverse current to ensure identical counting conditions. No statistically significant variations in the counting rates were observed throughout the experiment and in the control. This allowed us to place a 2σ upper bound for the average fusion rate of $< 2.1 \times 10^{-22}$ fusion neutrons per (dd pair) per sec. Similarly, no statistically significant variations were observed in the plastic scintillator counter throughout the active and background measuring periods. Table I summarizes data on counting rates for all detectors and experiments reported in this paper, while Table II shows upper limits for the various fusion rates calculated from the data.

B. "Twin" Pd cylinder cell

The data from the various detectors were analysed for statistically significant differences between cell A and cell B (H_2O, D_2O respectively). In particular, for the two Ge detectors we searched for statistically significant differences in the counting rates of the 2224-keV γ-ray in both detectors and none was found; in addition we searched the spectrum from the detector in setup-II for evidence of a 5.494-MeV

γ-ray from the possible d(p,^{3}He)γ fusion reaction. The γ-ray spectra in this region indicated no evidence for such a γ-ray throughout the experiment, and the counting rates in this energy region were the same for cells A and B (see Table I) in a 6-keV wide region centered at this energy. No efficiency calibration near this energy was performed, and therefore no absolute upper limit could be calculated for the d(p,^{3}He)γ fusion reaction.

Similarly, we searched for evidence of bremsstrahlung and Pd K_α X-rays from slowing down of possible charged particle fusion products in the X-ray detector. Again, no differences in the X-ray spectra and counting rates between the two cells were observed and an upper limit on the fusion-induced Pd K_α X-ray production rate was calculated and is included in Table II.

The NE213 liquid scintillator was run only in scaler mode gated by the neutron window of the PSD electronics. The threshold settings of the gate were checked every 24 hours during the exchange of the cells to ensure reliable n/γ discrimination. Only 7.5 days of data were collected, after which time the n/γ discrimination deteriorated due perhaps to poisoning of the scintillating liquid. The upper limit for the rate of fusion neutron production, calculated from the difference between cell A and B counting rates, is shown in Table II.

The neutron spectra taken with the ^{3}He spectrometer were analysed in the energy regions (1) near the thermal peak ($\sim 0 < E_n < 0.08$ MeV), (2) in the range $0.08 < E_n < 2.4$ MeV and (3) in the range $2.4 < E_n < 8.5$ MeV. In no single spectrum nor in the summed spectrum for a single cell over the entire measurement period was any indication found for a full-energy peak near 2.45 MeV. No qualitative differences were evident in the spectral distributions from the two cells (Fig.3, top). Besides the thermal peak, the spectrum is dominated by a broad distribution in the energy $2.9 < E_n < 4.4$ MeV. This is clearly associated with background and may be due in part to trace alpha-emitting impurities in the construction materials of the detector.

To check for the possibility of fusion induction due to mechanical stressing of the cathode, the current was cycled on and off when a cell was viewed by the ^{3}He detector. Following a long period of steady-state current (400 mA), five cycles of current off for 20 min and current on for 2 hrs were applied followed by recharging at the normal steady state. The cells were then relocated after a total period of 24 hours and the procedure repeated for the second cell. The entire sequence was repeated twice to obtain ten on/off cycles for each cell. Signals from the ^{3}He detector corresponding to neutron energies in the range 0–2.4 MeV were multi-scaled at a dwell time of 20s per channel and complete spectra recorded for individual parts of the stressing regimen.

A summary of the average count rates from both cells during the entire 25 days

of counting is given in Fig.4. Very significant variations in background rates (up to 20%) were experienced during this period (Fig.4, top) which emphasizes the importance of the "twin" cell procedure. No significant difference in count rates for the two cells over the first 12 days of measurement was found, and no trace of spectral characteristics from the interaction of 2.45-MeV neutrons was observed. This can be seen in Fig.3 (bottom), which represents the difference between summed spectra for the two cells (B−A) after normalizing by counting time.

During the period of stressing, the count rates of 4 cycles for cell B (D_2O) appear larger than those for cell A. However, the thermal peak from only one of these is significantly larger than the others (Fig.4, middle), and the ratio of its intensity to that in the energy range 2.4-3.6 MeV is the same, within errors, as that seen over the entire duration of the experiment (Fig.4, bottom). Since there is no contribution from 2.45-MeV neutrons in the high-energy region, this anomaly is clearly associated with an increase in electronic noise or general background. Furthermore, the difference between the summed spectra (B−A) over this period showed no significant residuals. While it is possible that some excess neutron intensity may be associated with stressing of the Pd electrode in the D_2O cell, our data are insufficient to rule out simple background fluctuations as the source. From the integral differences in the count rates during these measurements, upper limits for the production of fusion neutrons were calculated and are listed in Table II.

C. Thin Pd foil cell

A spectrum from the SSB detector of the thin Pd foil cell accumulated over 8.3 days, is shown in Fig.5. Energy calibration was obtained using ^{252}Cf, ^{237}Np and ^{243}Am α-sources. The expected spectral distribution from ^3He, tritons and protons produced by expected d-d fusion were simulated with the TRIM code of Ziegler[5]. Since the detector noise extends to about 700 keV, only the proton distribution could be used to search for the d(d,p)t branch.

This experiment is similar to the one reported recently be Ziegler et al.[3] Their gross counting rate in the 1–3 MeV region was $\sim 5.4 \times 10^{-5}$ s^{-1}, irrespective of the D loading of their Pd foil. Here we obtained $(5.1 \pm 1.8) \times 10^{-5}$ s^{-1} for H_2O and $(4.7 \pm 1.6) \times 10^{-5}$ s^{-1} for D_2O. By flushing our detector headspace with N_2 we decreased dramatically the count rate in the 3–8 MeV region due to α-emitting contaminants in the atmosphere, to a mean value of $(2.1 \pm 0.7) \times 10^{-5}$ s^{-1} MeV^{-1}, similar to our rates in the 1–3 MeV region. It has been suggested[6] that this residual count rate is due to α-emitting trace radioactive impurities in the Pd foil.

In Table II we include the 2σ upper limit for the rate of the d(d,p)t fusion branch calculated from our data. We have corrected for geometric collection efficiency and

the limited volume of the Pd foil which was sampled[6].

VI. SUMMARY AND CONCLUSIONS

For the series of experiments reported herein, careful attention was paid to experimental design, especially with regard to control experiments and calibration of equipment. A matrix of four conditions was studied whenever possible and appropriate: 1) light water electrolysis, Pd cathode, 2) light water electrolysis, Pd anode, 3) heavy water electrolysis, Pd cathode, and 4) heavy water electrolysis, Pd anode. In order to minimize bias, a "single blind" protocol was adopted at every opportunity so that experimenters did not know the identity of the sample which was being measured or analyzed.

The present series of experiments showed no positive evidence which supports the hypothesis that a nuclear fusion process took place in any of our deuterium-loaded palladium samples, either at steady state or in transient response to a variety of perturbations to the cells. From average count rates, upper bounds, many orders of magnitude below those implied by FP[1], and only slightly higher than the average rate reported by Jones[2], were placed on the fusion neutron production rates. The upper bound for a fusion proton production rate is an order of magnitude smaller than implied by Jones[2], assuming the normally expected d(d,p)t and d(d,n)^{3}He branching ratio.

Chemical analysis of our "twin" Pd cylinder electrolyte solution (Table III) revealed no measurable increase in tritium concentration, beyond the effects of electrolytic isotope enrichment during our experiment. A statistically-significant increase of 2.0 dpm $(mL)^{-1}$ would have corresponded to the production of $> 8 \times 10^8$ atoms of tritium, approximately the threshold for helium analysis, and to a fusion rate of 4×10^{-20} (dd pair)$^{-1}$ s^{-1}, well above our detection limit in all cases.

Because of relatively low detection efficiency, we cannot rule out the possibility of infrequent bursts of neutrons during stressing of the Pd cathodes. We note that some channels in our multi-scaled data contained counts greatly exceeding the average, but these could be readily associated with known noise sources and no trace of spectral effects from 2.45-MeV neutrons was found in any of our data. This emphasizes the importance of spectroscopic measurements in the presence of a large background. Finally, a means of obtaining simultaneous or near-simultaneous background measurements is crucial for avoidance of "false-positive" results because of the large fluctuations that are seen in the neutron background.

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[1]M. Fleischmann, S. Pons, and M.J. Hawkins **J. Electroanal. Chem. 261** (1989) 301 and erratum, **263** (1989) 187.

[2]S.E. Jones, E.P. Palmer, J.B. Czirr, D.L. Decker, G.L. Jensen, J.M. Thorne, and S.F. Taylor **Nature 338** (1989) 737.

[3]J.F. Ziegler, T.H. Zabel, J.J. Cuomo, V.A. Brusic, G.S. Cargill, III, E.J. O'Sullivan, and A.D. Marwick **Phys. Rev. Lett. 62** (1989) 2929.

[4]H. Franz, W. Rudolph, H. Ohm, K.-L. Kratz, G. Herrmann, F.M. Nuh, D.R. Slaughter, and S.G. Prussin **Nucl. Inst. Meth. 144**(1977) 253; W.C. Sailor, and S.G. Prussin **Nucl. Inst. Meth. 173**(1980) 511; H. Ohm, K.-L. Kratz, and S.G. Prussin **Nucl. Inst. Meth. Phys. Res. A256** (1987) 76; W.C. Sailor, and S.G. Prussin **Nucl. Inst. Meth. A270** (1988) 527; and references therein.

[5]"TRIM-86" Computer code described in: *The Stopping of Ions in Matter* , J.F. Ziegler ed., Pergamon Press, New York, 1985, vol. 1, ch. 8.

[6]P.B. Price, S.W. Barwick, W.T. Williams and J.D. Porter, **Phys. Rev. Lett.** (submitted 1989).

FIG. 1. Schematic layout of "twin" cell experiment. The area surrounding the ^{3}He n detector in Setup-II was not completely filled with H_2O bottles and paraffin wax blocks to prevent mechanical disruption of the detector. H_2O bottles and sheets of Cd were used to partially isolate the setups from each other.

FIG. 2. Electolytic cell with thin Pd foil electrode and Si surface barrier detector for the detection of charged particles.

FIG. 3. Top - the summed spectrum from the ^{3}He detector for cell B over the first 12 days of measurements in the "twin" cell experiment. The energy scale represents deposited energy based on Q = 0.764 MeV for the ^{3}He(n,p)^{3}H reaction. The spectral features expected from the 2.45-MeV neutron interactions include a full-energy peak at 3.21 MeV, a continuum in the range 0–2.45 MeV from neutron scattering on protons because of CH_4 in the gas mixture, a continuum in the range 0–1.65 MeV from elastic scattering on ^{3}He (upper limit reduced from the expected 1.85 MeV because of recombination effects), and a thermal peak centered at 0.764 MeV from scattering external to the detector. Bottom - the difference between the time-normalized summed spectra for cell B and cell A over the same measurement period. The net difference at low energies as well as the small differential distribution located at the midpoint of the thermal peak indicates a slight gain shift between the two spectra.

FIG. 4. ^{3}He detector count rates for neutron energies ~0–2.4 MeV (top) and in the thermal peak alone (middle) for the "twin" cells over the duration of the measurements. The ratios of thermal rates to the corresponding rates for neutron energies 2.4–3.6 MeV are given at the bottom. Energies shown are deposited energies based on Q = 0.764 MeV for the ^{3}He(n,p)^{3}H reaction. The period of time marked "O/O" corresponds to the experiment where the current was turned off for 20 min. and then turned on for 2 hours as described in the text. The period marked "BLANKS" corresponds to background counting with no electrolysis.

FIG. 5. Spectrum of the SSB detector. Total accumulation time 8.3 days. Pd foil cathode in 1.0 M $LiOD/D_2O$, operated at 33.8 mA/cm^2. 33 counts were observed in the $(1-3)$ MeV range characteristic of recoiling fusion protons. Over the same period of time, 36 counts were obtained using 1.0 M $LiOH/H_2O$ and the same operating conditions.

TABLE I. **Summary of Counting Rates**

Cell	Detector	Radiation	Counting Period (Days)	Counting Rate (cps)	Efficiency
		Pd Wire Experiment			
Exp.	Ge	2224-keV γ-ray	12.1	$5.5 \times 10^{-4} \pm 8.8\%$	1.51×10^{-4}
Control	Ge	2224-keV γ-ray	9.3	$5.0 \times 10^{-4} \pm 11.3\%$	1.51×10^{-4}
Exp.	Plastic Scintillator	Charged particles	11.6	$1.73 \pm 0.08\%$	–
Control	Plastic Scintillator	Charged particles	7.9	$1.75 \pm 0.09\%$	–
		"Twin" Pd Cylinder Experiment			
A	Ge Setup-I	2224-keV γ-ray	9.3	$4.5 \times 10^{-4} \pm 14.8\%$	1.51×10^{-4}
A	Ge Setup-II	2224-keV γ-ray	10.0	$5.9 \times 10^{-4} \pm 20.4\%$	1.10×10^{-4}
A	Ge Setup-II	5.494-MeV γ-ray	10.0	$2.02 \times 10^{-4} \pm 9.6\%$*	–
B	Ge Setup-I	2224-keV γ-ray	10.0	$4.5 \times 10^{-4} \pm 13.0\%$	1.51×10^{-4}
B	Ge Setup-II	2224-keV γ-ray	9.3	$4.3 \times 10^{-4} \pm 25.0\%$	1.10×10^{-4}
B	Ge Setup-II	5.494-MeV γ-ray	9.3	$1.58 \times 10^{-4} \pm 10.7\%$*	–
A	Ge X-ray	21.1-keV K_α X-ray	6.5	$0.429 \pm 1.4\%$*	2.4×10^{-3}
B	Ge X-ray	21.1-keV K_α X-ray	6.1	$0.424 \pm 1.5\%$*	2.4×10^{-3}
A	Plastic Scintillator	Cosmic ray, etc.	5.8	$0.23 \pm 0.3\%$	–
B	Plastic Scintillator	Cosmic ray, etc.	5.9	$0.23 \pm 0.3\%$	–
A	NE213 Liquid Scintillator	n> 1 MeV	3.7	$7.3 \times 10^{-3} \pm 2.1\%$	2.79×10^{-3}
B	NE213 Liquid Scintillator	n> 1 MeV	3.8	$7.1 \times 10^{-3} \pm 2.1\%$	2.79×10^{-3}
A	^{3}He	n_{th}	9.8	$5.85 \times 10^{-2} \pm 1.5\%$	1.30×10^{-2}
B	^{3}He	n_{th}	9.1	$5.84 \times 10^{-2} \pm 1.4\%$	1.30×10^{-2}
		Thin Pd Foil Experiment			
D_2O	SSB	Protons 1–3 MeV	8.3	$4.7 \times 10^{-5} \pm 17.4\%$	0.15
H_2O	SSB	Protons 1–3 MeV	8.2	$5.1 \times 10^{-5} \pm 16.7\%$	0.15

* No peak observed; rate is for continuum background in this energy region

TABLE II. **Limits on Cold Fusion Rates in Pd** †

Detector	Rate (d,d fusion)
	Pd Wire Experiment
Ge (n,γ)	$< 2.1 \times 10^{-22}$ n/(dd sec)
	"Twin" Pd Cylinder Experiment (Setup-I)
Ge (n,γ)	$< 1.3 \times 10^{-22}$ n/(dd sec)
NE213 LS	$< 1.7 \times 10^{-23}$ n/(dd sec)
Ge X-ray	$< 1.7 \times 10^{-21}$ X/(dd sec)*
	"Twin" Pd Cylinder Experiment (Setup-II)
Ge (n,γ)	$< 3.3 \times 10^{-22}$ n/(dd sec)
^{3}He n spec.	$< 2.1 \times 10^{-23}$ n/(dd sec)
	Thin Pd Foil Experiment
SSB	$< 5.9 \times 10^{-25}$ p/(dd sec)

†2σ limits

* Calculated from background rates in the Pd K_α region

TABLE III. **Analysis of water used in electrochemical cells**

	Stock		After Electrolysis	
	H_2O†	D_2O‡	H_2O	D_2O
^{1}H NMR* H (%)	–	0.13 ± 0.01	–	0.17 ± 0.01
^{2}H NMR* D (%)	0.014 ± 0.001	–	0.057 ± 0.0004	–
^{3}H LS T (dpm/mL)	< 2	17.6 ± 0.9	$< 2^a$,$< 2^b$	27.9 ± 0.8^a, 28.6 ± 1.0^b

* High Resolution pulsed NMR on Bruker AM-500

†Purified to 18 Mohm·cm on 4-stage milli-Q system

‡Lot F7962 Cambridge Isotopes Laboratory

[a]Neutralized with glacial acetic acid

[b]Distilled to dryness

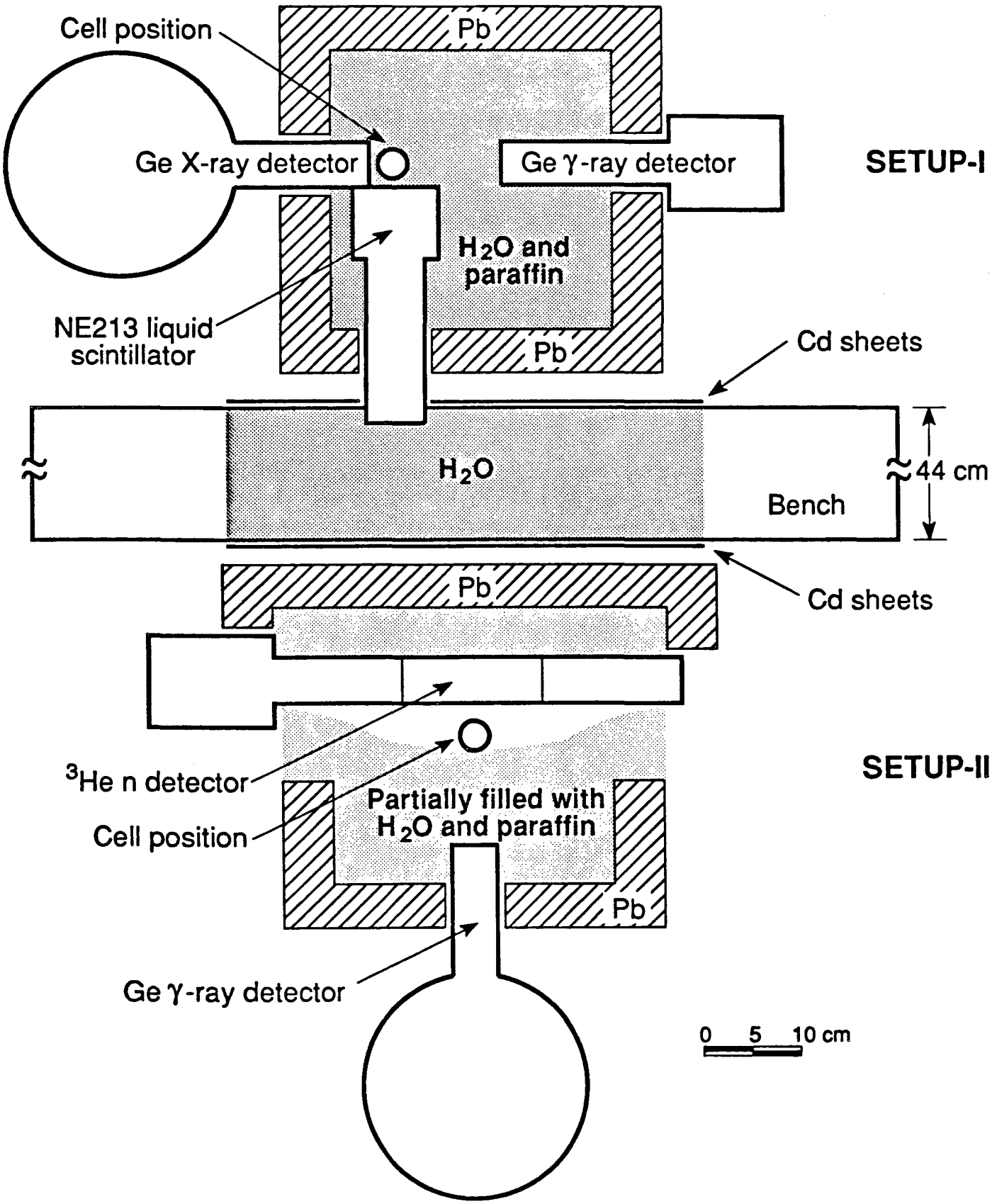


Fig. 1

XBL 897-6265

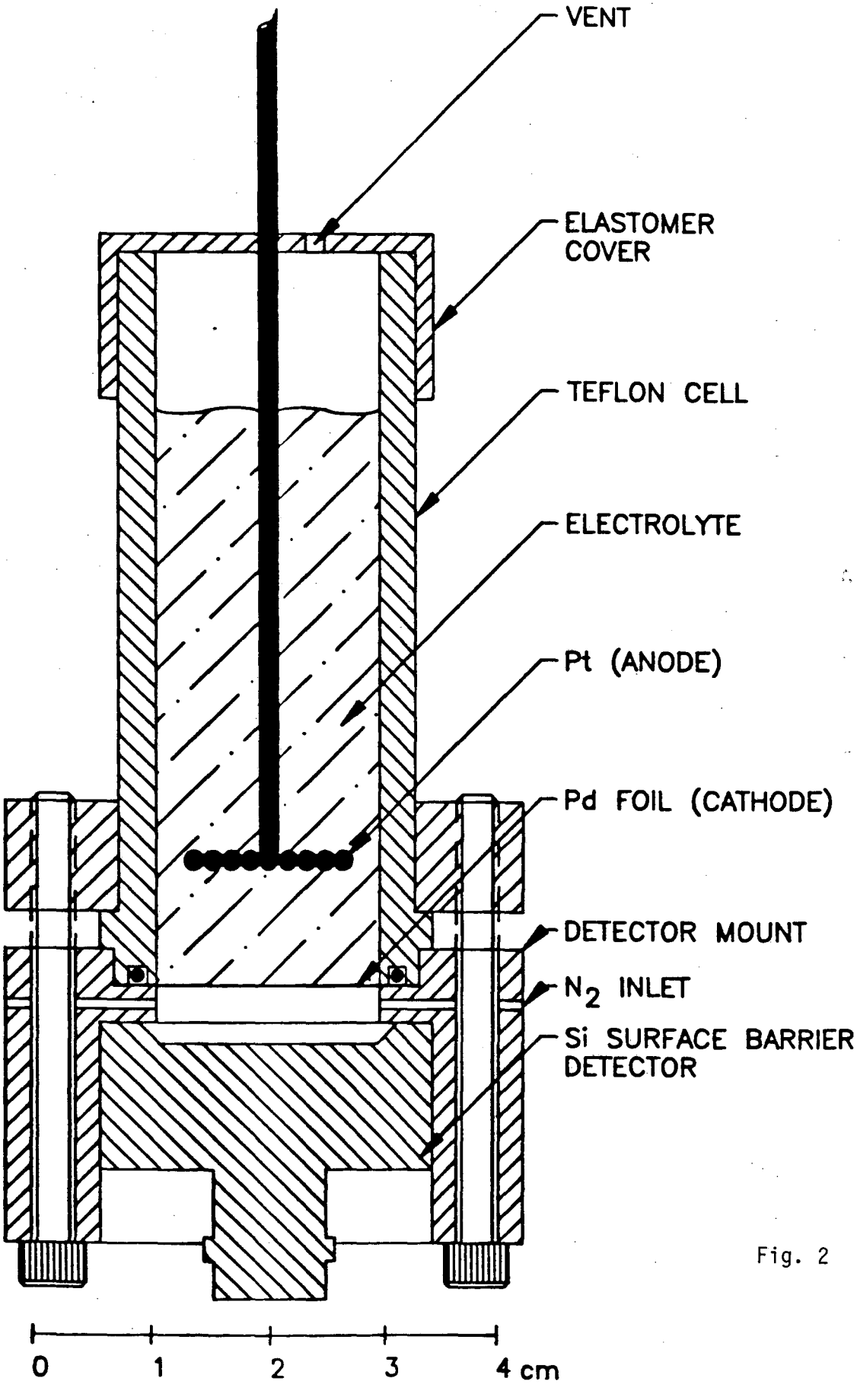

VENT
ELASTOMER COVER
TEFLON CELL
ELECTROLYTE
Pt (ANODE)
Pd FOIL (CATHODE)
DETECTOR MOUNT
N_2 INLET
Si SURFACE BARRIER DETECTOR
0
1
2
3
4 cm

Fig. 2

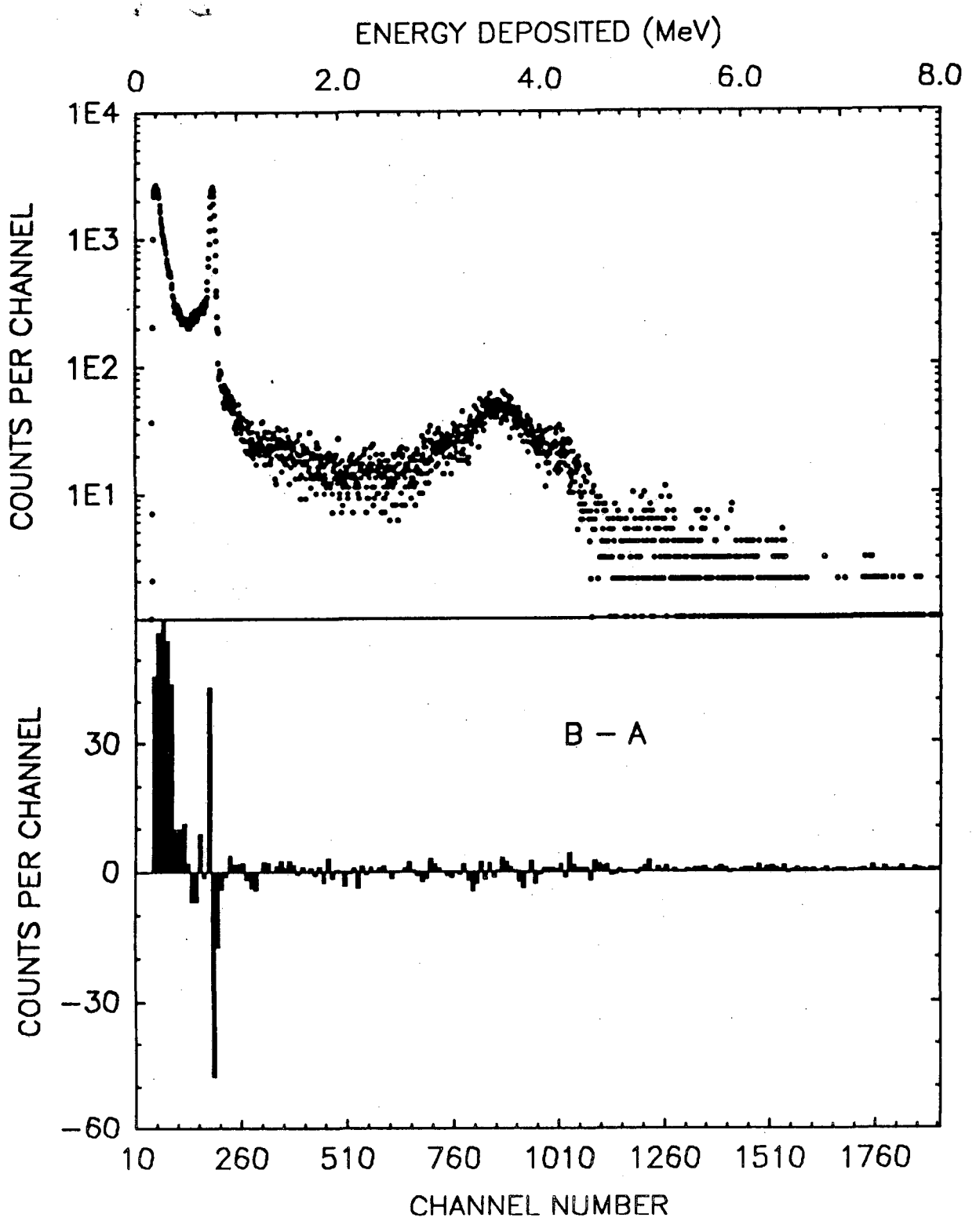
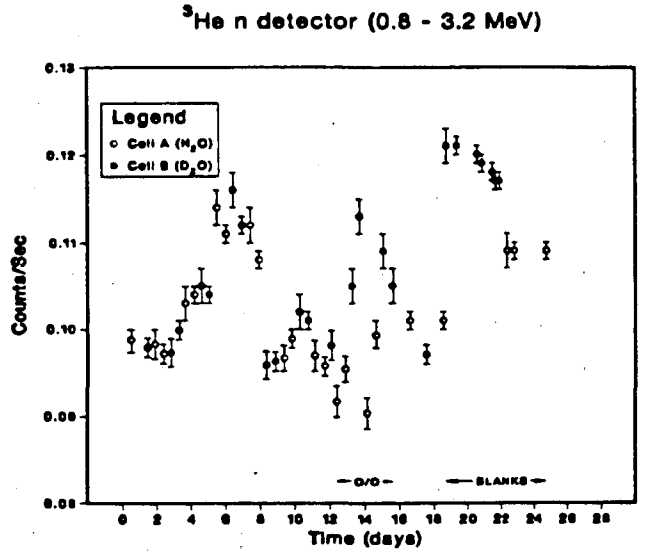
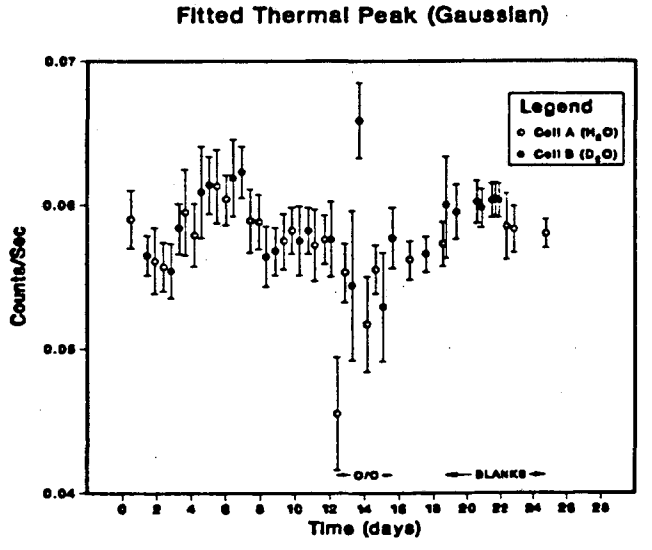

ENERGY DEPOSITED (MeV)
0
2.0
4.0
6.0
8.0
1E4
1E3
1E2
1E1
COUNTS PER CHANNEL
B – A
30
0
−30
−60
COUNTS PER CHANNEL
10
260
510
760
1010
1260
1510
1760
CHANNEL NUMBER

Fig. 3


^{3}He n detector (0.8 - 3.2 MeV)
Legend
Cell A (H_2O)
Cell B (D_2O)
Counts/Sec
Time (days)
O/O
BLANKS


Fitted Thermal Peak (Gaussian)
Legend
Cell A (H_2O)
Cell B (D_2O)
Counts/Sec
Time (days)
O/O
BLANKS

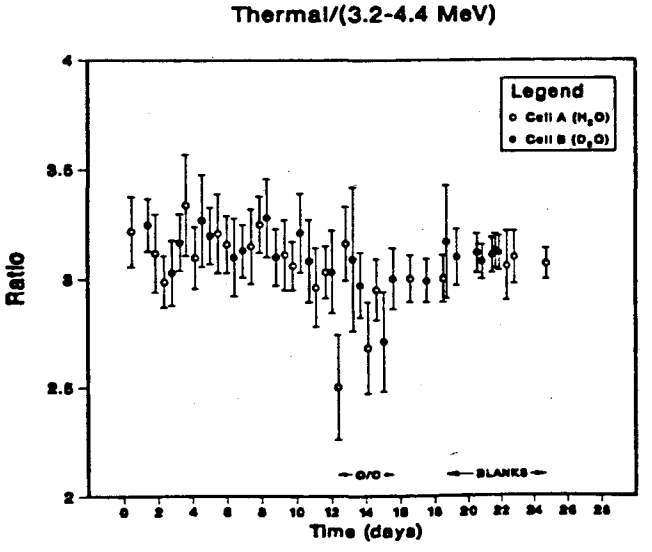

Thermal/(3.2-4.4 MeV)
Legend
Cell A (H_2O)
Cell B (D_2O)
Ratio
Time (days)
O/O
BLANKS

Fig. 4

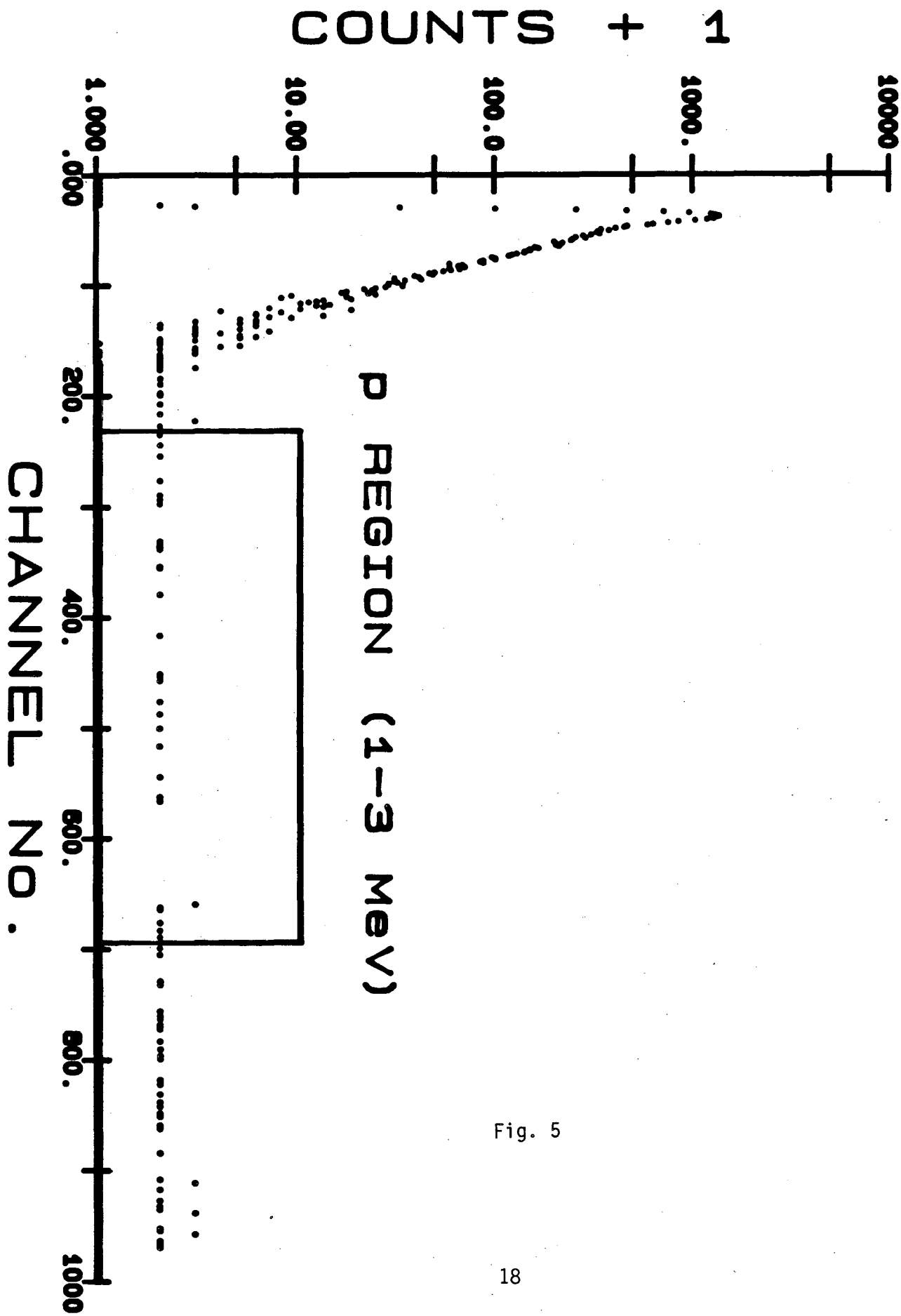

COUNTS + 1
1.000
10.00
100.0
1000.
10000
.000
200.
400.
600.
800.
1000
CHANNEL NO.
D REGION (1-3 MeV)

Fig. 5

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