Observation of High Multiplicity Bursts of Neutrons During Electrolysis of Heavy Water with Palladium Cathode Using the Dead-Time Filtering Technique

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Abstract

A series of experiments were carried out to detect production of neutrons from a commercial (Milton Roy) palladium-nickel electrolytic cell operated with 0.1 M LiOH or LiOD as the electrolyte at a current density of $\sim$ 80 mA/cm2. Neutron emission was monitored using a bank of 16 BF_3 detectors embedded in a cylindrical moderator assembly. A dead-time filtering technique was employed to detect the presence of neutron "bursts" if any and characterize the multiplicity distribution of such neutron bursts. It was found that with an operating Pd-D_2O cell located in the centre of the neutron detection set-up, the daily average neutron count rate increased by about 9% throughout a one month period, over the background value of $\sim$ 2386 counts/day indicating an average daily neutron production of $\sim$ 2220 neutrons/day by the cell. In addition analysis of the dead-time filtered counts data indicated that about 6.5% of these neutrons were emitted in the form of bursts of 20 to 100 neutrons each. On an average there were an additional 6 burst events per day during electrolysis with LiOD over the daily average background burst rate of 1.7 bursts/day. The frequency of occurrence of burst events as well as their multiplicity was significantly higher with D_2O + LiOD in the cell when compared with background runs as also light water "control" runs.

1. Introduction

At the first annual conference on cold fusion[1], the authors had reported the results of multiplicity distribution analysis of neutron counts obtained from a commercial Milton Roy electrolytic cell operated with 5M NaOD. For this, counts obtained in 20 ms intervals from a bank of BF_3 counters embedded in a slab of hydrogenous moderator was statistically analysed. It was found that about 10 to 25% of the neutrons produced were in the form of bunches of 400 to 600 neutrons, the balance following Poisson statistics i.e. neutrons emitted randomly one at a time. But since the efficiency of neutron detection in those measurements was only about 1%, the probability of detecting more than one neutron out of a bunch of simultaneously emitted neutrons was extremely small for bursts of < 100 neutrons. Subsequent attempts in 1991, with a newly procured Milton Roy cell and using pulse shape discrimination method in conjunction with a proton recoil counter (threshold < 10 - 2 n/s), did not yield any evidence of neutron production. In this paper we report on further work carried out to detect and characterize neutron emissions from a large cathode area Pd-D_2O Milton Roy electrolytic cell using a high efficiency annular neutron detection apparatus. A novel

dead-time filtering technique (1, 2) was employed this time to monitor and analyze burst neutron emission.

The experiments were carried over a period of two months of which the first 15 days were devoted to background runs i.e. without cell. Electrolysis with LiOD was performed for the next 30 days while LiOH was used in the last 15 days to serve as "control".

2. Neutron Detection Set-up

The neutron detection system[3] comprised of a bank of 16 numbers of 50 mm dia, 300 mm long BF_3 counters embedded in a cylindrical plexiglass moderator assembly which could accommodate the electrolytic cell at its centre. An instantaneous burst of neutrons released into such a system is temporally stretched to a few tens of μs duration in the moderator owing to the statistical time spread inherent in the slowing down process. The detectors were arranged to form three independent channels as shown in Fig. 1. The outputs, from each of the preamplifiers contained in shielded boxes, was fed to spectroscopic amplifiers. A suitable window was selected in the single channel analyzers (SCA) around the peak region of the pulse height spectrum of the amplifier output. The output pulses from the SCA were passed through a dead-time unit to distinguish bunches of pulses within 100 μs of each other, by generating an output corresponding to the first such pulse and rejecting all subsequent pulses arriving during the preset dead-time of 100 μs. The data of six channels, corresponding to direct and dead-time filtered pulses from each of the three detector segments, was counted over 5s intervals and stored continuously by means of a personal computer (PC) controlled scalar.

The neutron monitoring system had an overall detection efficiency of ~ 10% and an average background of ~ 0.048±0.002 counts/s (or ~ 172 counts/hr) which was more or less constant over a 15 day period. The preamplifiers and detector connections were hermetically sealed and specially designed to minimize electromagnetic disturbances. The entire set-up, which was located inside a "Faraday cage", did not register any spurious counts even under the high humidity conditions of the Bombay monsoon season.

3. Electrolytic Cell and Experimental Protocol

The Milton Roy electrolytic cell consists of 16 numbers of tubular palladium cathodes with a total surface area of ~ 300 cm^2. A pair of outer and inner nickel tubes serve as anodes. The power supply was used in a constant current (galvanostatic) mode. For each case, the cell was run at a small current density of 20 mA/cm^2 on the first day for "conditioning", followed by operation at about 40 - 45 mA/ cm^2 on the 2nd day. Thereafter the main electrolysis was carried out at a steady current of ~80 mA/ cm^2. To compensate for losses due to electrolysis or evaporation, D_2O or H_2O, as required, was made up every morning.

4. Results and Discussion

Although data from the three segments of counters were recorded separately, prior to analysis, the counts data from all the three channels were summed up. The data of individual segments were used only to check for internal consistency. For each day, the 5s counts were totalized from all the 10,000 intervals. The total direct channel counts for each day for (a) no cell (detector set-up only)

case; (b) electrolysis run using D_2O + LiOD and (c) electrolysis run using H_2O + LiOH are summarized in Tables l (a) to l (c). The tables also show the frequency distribution of 5s counts for multiplicities up to 8. The frequency distribution expected for a Poisson distribution corresponding to the average background of each experiment, is indicated as N_{Pd} at the bottom of the Tables for comparison. The total counts for each day, listed in the last column of the tables, are shown plotted in Fig. 2. It is clearly evident that the average count rate has shifted up significantly during electrolysis with LiOD + D_2O, relative to the no cell case. During the experiment with light water however, there is a large variation initially and subsequently the average count rate comes down towards background level by the end of the experiments. The multiplicity distribution of 5s counts, integrated and normalized for a 30 day experimental duration for the three cases, are shown plotted in Fig. 3. While the electrolysis experiments are seen to contain counts with multiplicity of up to 8, the background run has a maximum multiplicity of 5 only.

The characteristics of burst events, obtained from the difference of direct and dead-time filtered counts of a given absolute time interval, are summarized in Tables 2 (a) – 2 (c) for the three sets of experiments. The tables represent the multiplicity distribution of counts obtained in 100 μs intervals following every neutron pulse which managed to trigger the dead-time gate. The data for the case of electrolysis using D_2O+LiOD solution (Table 2(b)) again shows several instances of high multiplicity burst events. (Data have been appropriately normalised to account for the fact that the duration of the D_2O experiment was twice that of the other two cases). The effect is more apparent when plotted (see Fig. 4) as day-wise variation of total burst counts, for all the three experiments. It is assumed here that in one 5s interval only one burst occurs (as the overall frequency is very small in comparison to the total numbers of intervals). Fig. 5 shows a plot of the number of events having a given multiplicity of counts per burst in a 100 μs interval over a 30 day period. Here again it is clear that with D_2O cell and to a lesser extent with H_2O cell, events with multiplicities as high as 4 to 7 are recorded whereas with background (no cell case) maximum multiplicity of counts observed is only 3.

5. Summary and Conclusions

The total neutron counts per day with the D_2O cell was found to be consistently ~ 9% above the background level. However in the case of the H_2O experiment, which was conducted immediately after a month long D_2O run, the average daily count rate was found to steadily decrease to background level (Fig. 2), suggesting that this behaviour can probably be attributed to the slow replacement of D by H, within the Pd cathodes over several days. The frequency distribution of 5s counts was close to Poisson distribution in case of background but contained several large multiplicity events in presence of the H_2O or D_2O cells (Fig. 3). Moreover while the background counts did not show even a single count with multiplicity of 4 or more throughout the 15 day period (Table 2(a)), there were several events with multiplicity of 6 and even 7 counts in the 100 μs duration data, in presence of H_2O or D_2O cells. On the whole the number of burst events were however very few, the average values being 1.7, 3.8 and 7.6 bursts per day for the cases of background, H_2O cell and D_2O cell respectively.

In the present experiment since the overall neutron detection efficiency was ~ 10%, one can say that approximately 10 neutrons are emitted by the electrolytic cell for every neutron detected. Likewise a multiplicity of 4 counts during a 100 μs interval implies emission of a burst of roughly 40 neutrons by the cell. Out of the 2608 neutrons detected per day in presence of the Pd-D_2O cell, after subtracting the background of 2386, the balance of 222 counts/day can be attributed to the cell. Of

this about 14.5 counts per day (see Tables 2 (a) and 2 (b)) can be accounted for by high multiplicity (>20) burst neutron emission. Thus the conclusion from the present series of experiments is that about 6.5% of the neutrons produced by the Milton Roy electrolytic cell can be attributed to high multiplicity (> 20 neutrons/burst) events, and the balance 93.5% is produced either as single neutrons (with Poisson distribution) or with multiplicity of < 20 neutrons.

The present experiment thus once again confirms that a small component of the neutrons emitted by $Pd-D_2O$ cells is produced in the form of temporally bunched neutrons. Any theoretical explanation of cold fusion must account for this phenomenon also.

References

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2. Degwekar, D.G. and Srinivasan, M., "A simple dead time method for measuring the fraction of bunched neutronic emission in cold fusion experiments", Ann. Nucl. Energy 17, 583 (1990).
3. Shyam, A., et. al, "Technique to measure small burst of neutrons in presence of background of other radiations and electromagnetic interference", Ind. J. Appl. Phys. **32,** 837 (1994).
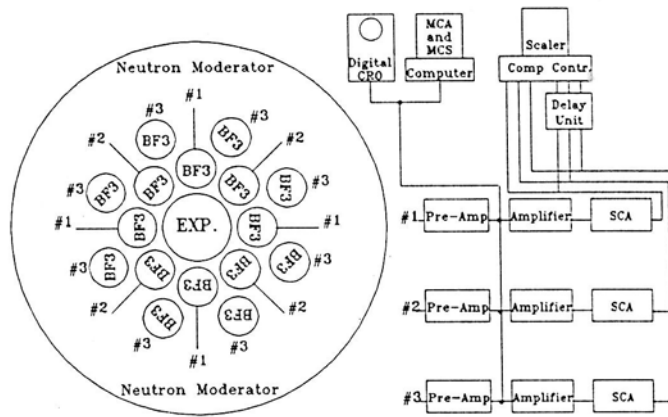
Figure 1. Schematic of Neutron Detection Setup

Train of pulses, as received from detector

Gate pulses provided by the Dead Time Unit (DTU)

Dead Time | Dead Time | Dead Time | Dead Time

Pulses after gating from Dead Time Unit

After Subtraction from the original train of pulses

No Burst | Burst of Two | Burst of One | No Burst
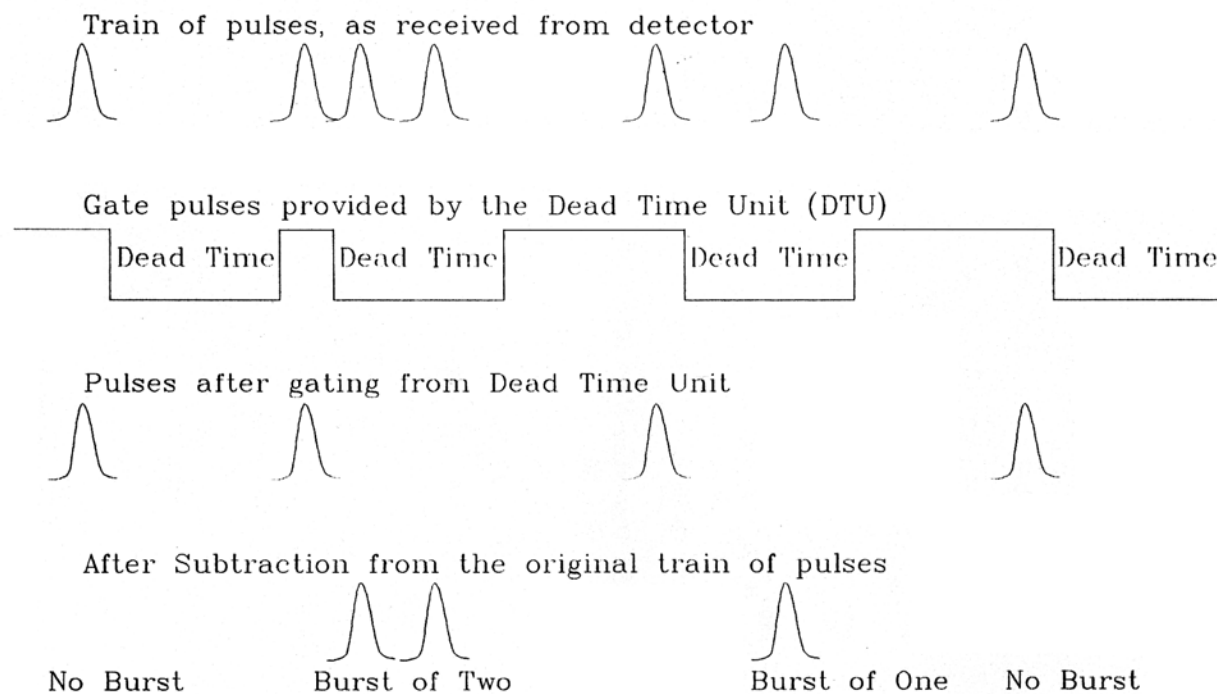
Figure 2. Principle of Burst Neutron Detection Using a Dead-Time Filter
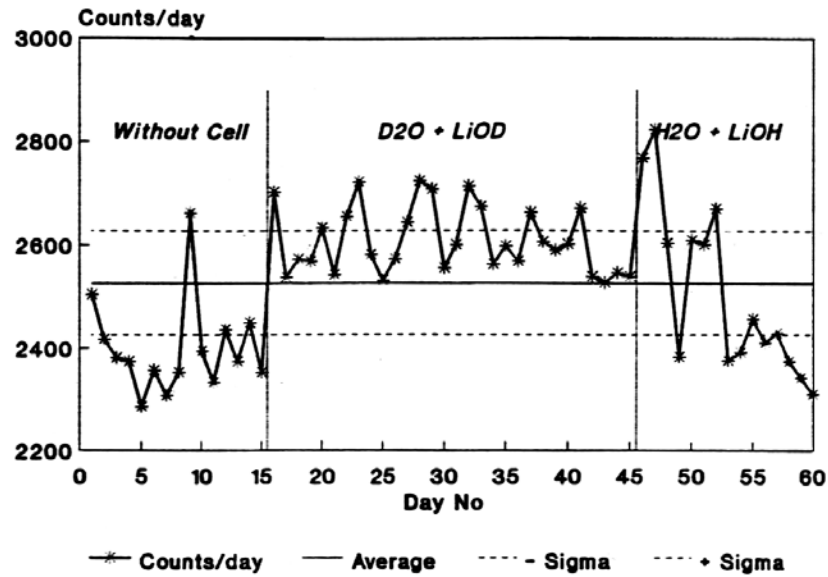
Figure 3. Variation of Total Direct Channel Counts
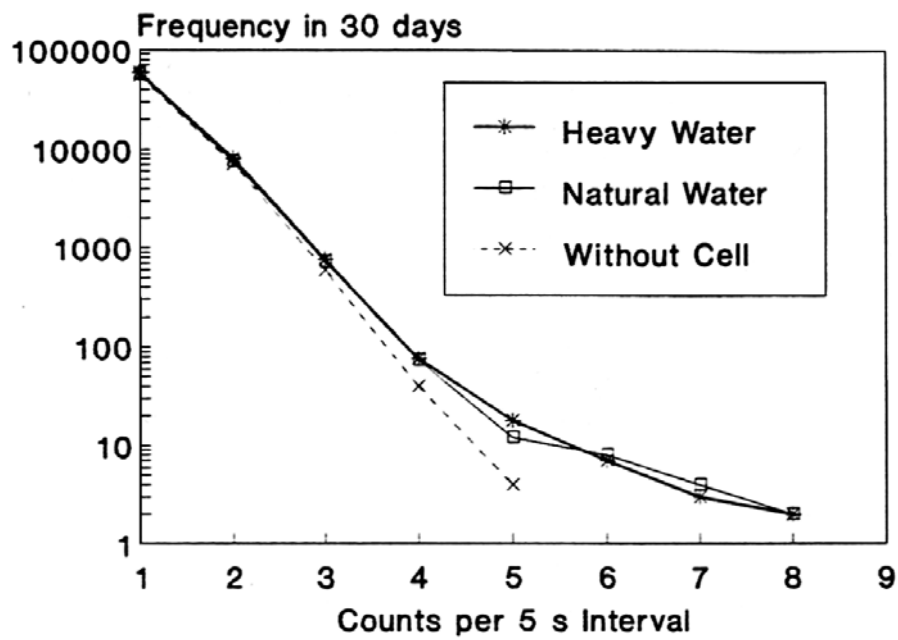
Figure 4. Frequency Distribution of 5 Sec Counts Integrated (Normalised) Over 30 Day Period
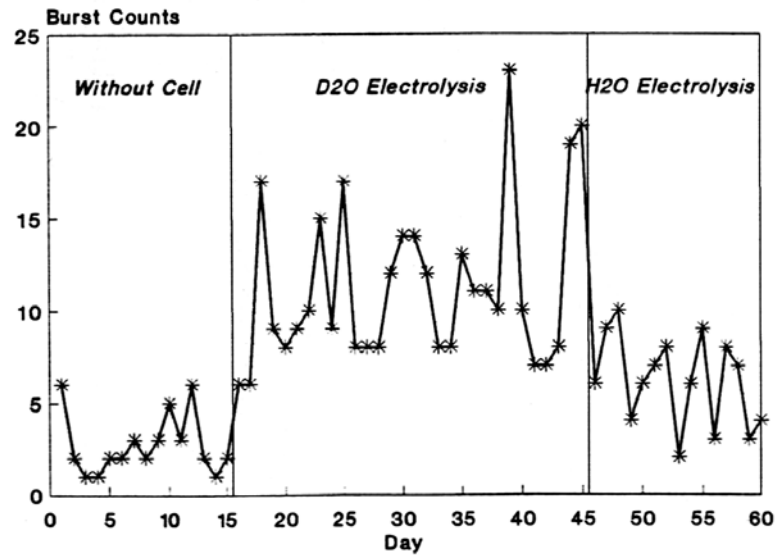
Figure 5. Day-wise Variation of Total Burst Counts
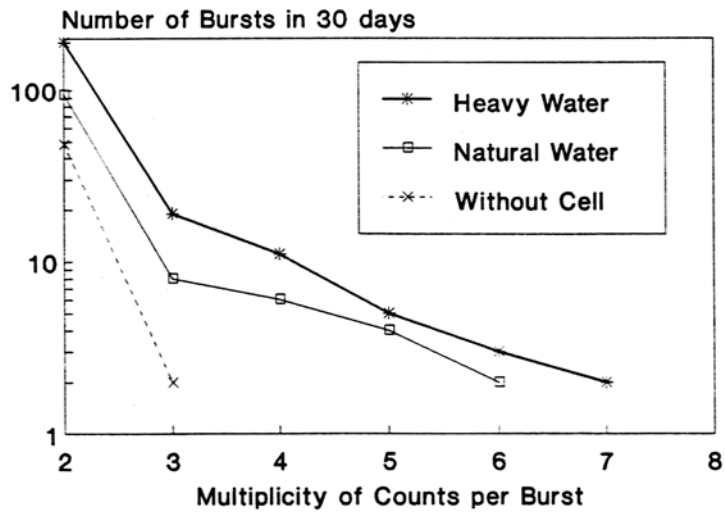
Figure 6. Frequency Distribution of Burst Counts Integrated (Normalised) Over 30 Day Period

Table 1. Direct Channel 5 sec Counts Data

1 (a): Background (No Cell) Case

	Frequency for Counts in 5 Second Intervals								
Day	1	2	3	4	5	6	7	8	Net
1	2001	228	16	0	0	0	0	0	2505
2	1877	231	25	1	0	0	0	0	2418
3	1864	222	23	1	0	0	0	0	2381
4	1808	245	20	4	0	0	0	0	2374
5	1836	195	17	1	1	0	0	0	2286
6	1800	257	13	1	0	0	0	0	2357
7	1793	222	21	2	0	0	0	0	2308
8	1816	249	10	2	0	0	0	0	2352
9	2012	280	30	0	0	0	0	0	2662
10	1847	236	21	2	1	0	0	0	2395
11	1822	229	18	0	0	0	0	0	2334
12	1883	243	20	2	0	0	0	0	2437
13	1873	220	19	1	0	0	0	0	2374
14	1895	244	21	1	0	0	0	0	2450
15	1876	207	18	2	0	0	0	0	2352
Tot	27803	3508	292	20	2	0	0	0	35985
N_{Pd}	28053	3415	277	16.9	0.82	0.033	0.001	$<10^{-4}$	

1 (b): With D_2O + LiOD

| Day | Frequency For Counts in 5 Second Intervals | | | | | | | | |
	1	2	3	4	5	6	7	8	Total
1	2049	280	27	2	1	0	0	0	2703
2	1948	254	26	1	0	0	0	0	2538
3	1978	255	24	3	0	0	0	0	2572
4	1973	259	22	3	0	0	0	0	2569
5	2029	261	26	1	0	0	0	0	2633
6	1981	235	28	2	0	0	0	0	2543
7	2037	255	22	9	0	1	0	0	2655
8	2051	277	31	3	0	2	0	0	2722
9	1970	255	31	2	0	0	0	0	2581
10	1982	237	21	3	0	0	0	0	2531
11	1970	256	18	2	3	1	1	0	2572
12	1980	275	28	5	2	0	0	0	2644
13	1986	331	24	1	0	0	0	0	2724
14	2029	284	32	0	3	0	0	0	2708
15	1996	245	19	2	1	0	0	0	2556
16	1976	269	22	2	1	0	0	1	2601
17	2052	276	29	1	1	1	1	0	2713
18	1996	287	28	5	0	0	0	0	2674
19	1957	273	17	1	1	0	0	0	2563
20	1974	257	32	2	0	1	0	0	2598
21	1982	248	22	6	0	0	0	0	2568
22	2009	274	30	4	0	0	0	0	2663
23	2041	262	12	0	1	0	0	0	2606
24	1930	278	32	2	0	0	0	0	2590
25	1987	261	27	3	0	0	0	0	2602
26	1983	278	29	4	3	1	0	1	2671
27	1937	269	20	1	0	0	0	0	2539
28	1938	243	28	5	0	0	0	0	2528
29	1935	266	25	1	0	0	0	0	2546
30	1914	277	20	1	1	0	1	0	2540
Tot.	59570	7977	752	77	18	7	3	2	78253
N_{Pd}	59966	7981	708	47.1	2.51	0.113	0.004	$<10^{-4}$	

1 (c): With H_2O + LiOD

				Frequency For Counts in 5 Second Intervals					
Day	1	2	3	4	5	6	7	8	Net
1	2082	302	26	0	1	0	0	0	2769
2	2100	301	32	4	2	0	0	0	2824
3	1996	242	35	3	0	0	1	0	2604
4	1836	246	17	1	0	0	0	0	2383
5	2020	249	31	0	0	0	0	0	2611
6	1930	285	30	3	0	0	0	0	2602
7	2024	271	27	6	0	0	0	0	2671
8	1894	208	18	3	0	0	0	0	2376
9	1836	226	22	6	0	1	0	1	2392
10	1824	277	23	1	0	1	0	0	2457
11	1895	223	22	1	0	0	0	0	2411
12	1913	220	19	3	0	0	1	0	2429
13	1867	223	17	1	0	1	0	0	2374
14	1822	225	21	2	0	0	0	0	2343
15	1839	195	16	3	3	1	0	0	2310
Tot.	28878	3693	356	37	6	4	2	1	37556
N_{Pd}	29089	3716	316	20.2	1.033	0.044	0.002	$<10^{-4}$	

Table 2. Burst Counts Data for 100 µs Intervals

2 (a): Only Counters, No Cell

Frequency for Burst Counts							
Day	**2**	**3**	**4**	**5**	**6**	**7**	**Total**
1	5	0	0	0	0	0	10
2	1	0	0	0	0	0	2
3	0	0	0	0	0	0	0
4	0	0	0	0	0	0	0
5	1	0	0	0	0	0	2
6	1	0	0	0	0	0	2
7	2	0	0	0	0	0	4
8	1	0	0	0	0	0	2
9	2	0	0	0	0	0	4
10	4	0	0	0	0	0	8
11	2	0	0	0	0	0	4
12	3	1	0	0	0	0	9
13	1	0	0	0	0	0	2
14	0	0	0	0	0	0	0
15	1	0	0	0	0	0	2
Tot.	**24**	**1**	**0**	**0**	**0**	**0**	**51**

Total Number of Bursts   **25**
Average Number of Bursts/day   **1.7**

2 (b): With D_2O + LiOD

	Frequency For Burst Counts						
Day	**2**	**3**	**4**	**5**	**6**	**7**	**Total**
1	3	1	0	0	0	0	9
2	5	0	0	0	0	0	10
3	3	2	1	0	0	1	23
4	6	1	0	0	0	0	15
5	7	0	0	0	0	0	14
6	8	0	0	0	0	0	16
7	3	0	2	0	0	0	14
8	3	0	2	0	1	0	20
9	8	0	0	0	0	0	16
10	6	1	0	2	0	0	20
11	5	1	0	0	0	0	13
12	7	0	0	0	0	0	14
13	4	0	1	0	0	0	12
14	11	0	0	0	0	0	22
15	9	0	0	1	0	0	23
16	5	2	0	1	0	0	21
17	6	0	0	0	1	0	18
18	5	1	0	0	0	0	13
19	5	1	0	0	0	0	13
20	10	1	0	0	0	0	23
21	4	3	0	0	0	0	17
22	8	1	0	0	0	0	19
23	4	1	1	0	0	0	15
24	8	1	1	1	1	0	34
25	6	0	1	0	0	0	16
26	6	0	0	0	0	0	12
27	6	0	0	0	0	0	12
28	4	0	1	0	0	0	12
29	13	1	1	0	0	0	33
30	11	1	0	0	0	1	25
Tot.	**189**	**19**	**11**	**5**	**3**	**2**	**529**

Total Number of Bursts   **229**
Average Number of Bursts/day   **7.6**

2 (c): With H_2O + LiOD

	Frequency for Burst Counts						
Day	**2**	**3**	**4**	**5**	**6**	**7**	**Total**
1	1	0	0	1	0	0	7
2	8	0	0	0	0	0	16
3	6	0	1	0	0	0	16
4	3	0	0	0	0	0	6
5	3	1	0	0	0	0	9
6	6	0	0	0	0	0	12
7	5	1	0	0	0	0	13
8	1	0	0	0	0	0	2
9	5	0	0	0	0	0	10
10	1	1	0	0	1	0	11
11	2	0	0	0	0	0	4
12	0	1	1	1	0	0	9
13	3	0	1	0	0	0	10
14	2	0	0	0	0	0	4
15	1	1	0	0	0	0	5
Tot.	**47**	**4**	**3**	**2**	**1**	**0**	**134**

Total Number of Bursts   57
Average Number of Bursts/day   3.8