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Upper limit for neutron emission from cold *d-t* fusion

J. R. Southon,*† J. W. Stark, J. S. Vogel,*† and J. C. Waddington
Tandem Accelerator Laboratory, McMaster University, Hamilton, Ontario, Canada L8S 4K1
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Reports that fusion of hydrogen isotopes may occur at room temperature during the electrolysis of D_2O led us to investigate possible *d-t* fusion reactions in an electrolysis cell with a tritiated Ti cathode. A search for 14-MeV neutrons from this reaction, using an NE213 scintillator detector and pulse-shape discrimination techniques, showed no significant deviations from background levels. We derive an upper limit of 10^{-23} s^{-1} for the fusion rate per *d-t* pair.

Reports suggesting that cold *d-d* fusion may occur at measurable rates in Pd or Ti cathodes during electrolysis of D_2O (Refs. 1 and 2) led us to consider whether other cold fusion reactions could also take place. There is great interest in the *d-t* reaction at keV energies as a source of fusion power, as the energy release and the reaction cross section are both high. Calculations[3,4] predict that this process is severely inhibited relative to *d-d* at room temperature, but the *d-d* fusion rates predicted by those calculations disagreed with the initial reported observations[1,2] by factors of 10^{30}. Therefore, when this experiment was undertaken, the possibility of measurable *d-t* rates could not be excluded. Subsequent work[5-8] failed to confirm the high *d-d* fusion rates reported earlier,[1,2] but experimental investigations of the *d-t* system remain of interest. Hydrogen isotopes remain stably absorbed in Ti at room temperature, and electrolysis of D_2O using a tritiated Ti cathode provided a simple means of carrying out these investigations with little radiological hazard.

The experiment was setup in a tritium fume hood in the basement of the McMaster Tandem Accelerator Laboratory, in a location shielded overhead by 80 cm of concrete. All of the electronics were operated from the laboratory "clean" power circuits, and an isolating transformer was used to avoid ground loop noise and to act as a choke in reducing fast line transients. Data were accumulated only when the accelerator was off, and most runs were taken during the electrically quieter night hours.

The electrochemical cell was contained in an uncapped 2 cm diameter test tube filled with electrolyte to a depth of 7–8 cm. The anode for the cell was a spiral of several turns of 1 mm Pt wire filling the bottom 5 cm of the test tube. The cathode consisted of a $15\times0.4\times0.05$ cm strip of Ti, with one end preloaded with tritium, positioned axially in the tube. The tritium was loaded by heating the strip with a rf heater and allowing it to cool under several hundred torr of tritium gas, after the Ti had been etched with HF and outgassed by repeated heating under vacuum. The quantity of tritium absorbed was determined from the pressure drop in the loading cell: approximately 1.7×10^{21} atoms were absorbed into about 3 cm of the strip, for a *t*/Ti ratio $\sim$0.5. We made no attempt to increase this ratio further, as the material becomes very brittle. As expected, the tritium was well contained by the Ti, and tritium levels measured around the apparatus during the experiment were close to background.

The electrolyte used in the cell was D_2O (>99.8% D), to which was added sufficient Li_2CO_3 to raise the Li^+ concentration to 0.1*M*, and HNO_3 to bring the *p*H to 2.5. Lithium was chosen to avoid problems of "poisoning" of the cathode by reduction of cations which may have occurred in earlier work.[1] The cell voltage and current measured at the power supply were typically 4.5 V and 300 mA, respectively, and D_2O losses were about 1 cm^3/day. Since these losses were made up with electrolyte, the actual Li^+ concentration varied between about 0.1*M* and 0.15*M* over the course of the experiment.

Neutrons were detected with a 5.1×5.1 cm NE213 scintillator coupled to an RCA 8850 photomultiplier, with the front face of the detector 1.5 cm from the cathode. Separation of neutron and gamma pulses was implemented by measuring dynode pulse rise times to produce a pulse-shape spectrum, using a fast leading edge and constant fraction timing. Since the differences in the scintillator response to neutrons and gammas are in the slow component of the light output,[9] the constant fraction unit was set up to measure the time taken to collect a high fraction (80%) of the total charge from the photomultiplier.

The scintillator response was calibrated using standard gamma sources, and an Am-Be neutron source which provides gammas and neutrons of up to 4.4 and 11 MeV, respectively. The response for 11 MeV neutrons was roughly equivalent to that from gamma rays of 5.7 MeV, close to results obtained previously[10] for NE213. A lower level pulse-height cutoff equivalent to the maximum pulse height from a 1 MeV gamma (or 3 MeV neutron, Ref. 10) was adopted to reduce backgrounds and to ensure that *d-d* neutrons were not measured. The window for acceptable neutron events in the pulse-shape spectrum was set well to the neutron side of the valley between the neutron and gamma peaks, so that good discrimination against gammas would be maintained even in the event of drifts in the electronics. This resulted in a loss of between 3% and 5% of the neutron pulses above the pulse-height cutoff. The gamma peak-to-valley ratio in the Am-Be pulse-shape spectrum for events above the pulse-height cutoff was 60–70:1, which compares well with previous results.[10]

The experiment was carried out over a five day period. Runs were typically started during the afternoon or evening, run for 10–15 h, and terminated before the tandem accelerator was started up in the early hours of the morn-

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ing. Pulse-height spectra and arrival times (i.e., multiscaling spectra) for events satisfying the pulse-height and pulse-shape constraints were recorded using Tracor TN1705 and 1710 multichannel analyzers. Pulse-shape spectra from the Am-Be source were taken before and after every run to check for electronic drifts.

The results are summarized in Table I. The initial background (bkgd) runs 1 and 2 were taken with no cathode in the cell. The tritiated cathode was installed and the cell was turned on about 20 min before data taking for run 3 began. The cell was left on during the 14 h hiatus between runs 3 and 4 while the tandem accelerator was operated. The final background run was carried out with the cathode still in place, but with the cell switched off.

The results showed no significant differences in neutron rates between the background runs and those with the cell operating. The multiscaling spectra showed no evidence of bursts of neutrons beyond the normal variations expected from the average detection rates, and the pulse-height distributions for the *d-t* and background runs appeared identical. The mean neutron detection rates for the background and *d-t* runs are $(7.8\pm0.8)\times10^{-4}$ s^{-1} and $(7.2\pm0.9)\times10^{-4}$ s^{-1}, respectively. The difference is $(-0.6\pm1.2)\times10^{-4}$ s^{-1}, and we take the upper limit for *d-t* neutron detection to be 2.4×10^{-4} s^{-1} at two standard deviations. It is possible that the reaction turned on and off between runs 3 and 4, or that a "charging" period longer than two days is required before the reaction starts. However, this seems unlikely on the basis of the claim by Jones *et al.*[1] that any charging time for *d-d* is as short as 1 h.

An approximate efficiency calculation carried out for an on-axis point source of 14 MeV neutrons 1.5 cm from the scintillator, using an (n,p) scattering cross section of 750 mb,[11] yielded a detector efficiency of 2.5%. Taking into account the efficiency reduction from the extended nature of the tritiated cathode and the loss of events falling outside the energy and pulse-shape windows, we estimate the actual efficiency to be 1.3 ± 0.4%.

We measured the hydrogen uptake of an initially hydrogen-free cathode strip under electrolysis in 0.1*M* LiOH in conditions similar to those in the *d-t* experiment, by periodically removing it from the cell and weighing it after cleaning with trichlorethylene and distilled water.

TABLE I. Experimental runs and neutron count rates.

Run	Start time		Run time (h)	Neutrons detected	Count rate (10^{-4}s^{-1})
1 (bkgd)	12:00	(5/15/89)	14.7	44	8.3 ± 1.3
2 (bkgd)	10:36	(5/16/89)	4.3	18	11.8 ± 2.8
3 (*d-t*)	15:43	(5/16/89)	13.4	30	6.2 ± 1.1
4 (*d-t*)	19:29	(5/17/89)	10.0	31	8.6 ± 1.5
5 (bkgd)	17:04	(5/18/89)	12.1	25	5.7 ± 1.1

The cathode gained 12.1 mg over the first 25 h and an additional 1.4 mg over the next 75 h, corresponding to mean compositions in the 3.5 cm wetted length of the strip of $TiH_{1.8}$ and TiH_2, respectively, assuming that all of the additional mass was hydrogen. Though not definitive, these data suggest that the assumption of essentially complete hydrogen loading is a reasonable one for our experimental geometry, as do the results of Williams *et al.*,[5] who observed deuteride penetration rates of ~30 μm/h in Ti under similar conditions of electrolysis.

Assuming similar penetration of deuterium into the partially tritiated Ti lattice in our *d-t* experiment, we take the number of *d-t* pairs to be 1.7×10^{21}, determined by the quantity of absorbed tritium. The upper limit for the fusion rate per *d-t* pair calculated from the observed neutron flux is then about 10^{-23} s^{-1}. It should be noted, however, that attempts[5-8] to duplicate the earlier *d-d* experiments[1,2] revealed no statistically significant neutron fluxes above background. Either the earlier results are in error, or reaction rates for cold fusion processes (including *d-t*) may depend critically on subtle differences in experimental conditions.

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*Also at Archaeology Department, Simon Fraser University, Burnaby, British Columbia, Canada V5A 1S6.

†Present address: Center for Accelerator Mass Spectrometry, Lawrence Livermore National Laboratory, Livermore, CA 94550.

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