

POSSIBLE DEUTERIUM PRODUCTION FROM LIGHT WATER
EXCESS ENTHALPY EXPERIMENTS USING NICKEL CATHODES

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ABSTRACT

Nickel cathodes that produced excess enthalpy during the electrolysis of light water solutions were examined several weeks later (metachronously) by electron emission spectroscopy. The absence of an obvious significant emitting ash might be consistent with a short-lived product, loss of the product, self-absorption, or low signal to noise, or a new fusion pathway leading to a stable ash. The latter may contribute to the excess heat as well as other researchers' reported transmutation reactions through a portion of the pathway leading to the production of *de novo* deuterons. If deuterium is a product of these reactions then its production levels, the inevitable separation factors, and present detection thresholds pose a significant challenge for its unambiguous identification.

INTRODUCTION -- EXPLANATIONS NEEDED

The deuteron is rarely, if ever, considered a possible product of cold fusion systems. Although examination of a large subset of the cold fusion literature (>840 abstracts [1]) revealed eighteen papers concerning tritium or triton production [2-19], a similar search revealed none discussing deuterium or deuteron production. This absence exists even though a pathway to its production might also explain the excess heat [20-27] and reported transmutations [3,25,26,28-30]. There must be a reasonable explanation for the excess heat and transmutation reactions. In light water nickel cold fusion systems, even purported transmuted products do not appear to be proportional to the expected ash required for the observed excess enthalpy. The anomalous branching ratio remains as problematic as the excess heat, and the lack of a theoretical model has been a major cause of the knee-jerk dismissal of this scientific field.

The coulomb barrier approach has been examined by many hypotheses but screening factors alone may not be sufficient to explain the observations, and thus the reaction pathway probably does not involve the classic collision of two particles. Several hypotheses exist including "shrunken" hydrogen [20], positron emission [31], and possible deuteron production [2]. In the case of helium produced from deuterium-loaded palladium, the Phuson theory [33] has been developed describing the multi-body reactions involving the loaded deuterons within the lattice of the fully loaded Group VIII transition metal electrode cathodically driven to ~1 kilomolar electron density. In the Phuson theory, the phonon clusters distribute energy between the excited state and the ground state, thereby conserving momentum within the palladium.

Because there does not exist a similar theory for nickel, due to the excess heat observed and the lack of a clear electron emission in these metachronous experiments, we elected to look more closely at deuterium, its production, its spin structure, and its possible role in these phenomena. This paper introduces a pathway that could yield *de novo* deuterons and explain the observed transmutation reactions. The hypothesis suggests that

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energy may be transferred to the lattice through a phonon cloud (Phuson) as a non-bound virtual deuteron state collapses to the ground state configuration.

BACKGROUND -- DEUTERON STRUCTURE AND SEPARATION

Chemically, the hydrogen isotopes slightly differ in their equilibrium constants, enabling the difficult separation of deuterium oxide from ordinary water (Table 1). Although hydrogen was isolated as a distinct substance in 1766 by Cavendish, deuterium was separated electrolytically in 1932 by Urey, Brickwedde and Murphy after its spectroscopic identification one year earlier. The actual situation is complicated because of tritium and the fractional species (e.g., DTO). The fragile deuteron decomposes at temperatures cooler than our sun's core (~million degrees Kelvin). So even though interstellar clouds and protostars contain deuterium (abundance D/H = $\sim 1.9 \{+/-0.5\} 10^{-4}$) evolved stars have essentially none [34-36]. Therefore, deuterium oxide (D₂O), although ubiquitous (D/H = $\sim 1.5 10^{-4}$) *in situ* within ordinary water, must be commercially refined and purified at considerable difficulty to obtain the several megaliters per year required for its use as a moderator for some nuclear reactors. The difficulty in refining deuterium arises for several reasons. The separation of the deuterium is highly endothermic, and most of the separation factors are small (Fig. 1). Deuterium is generally refined electrolytically following cascaded refiners using the slight difference in the free energies of formation of different hydrogen, nitrogen, or sulfur compounds (e.g., ammonia-hydrogen exchange and the Girdler-Sulfide system) [37].

In a deuteron, the constituent neutron and proton have an exceptionally large internucleon separation. The importance is the deuteron binding energy is only ~ 2.2 MeV in a well of about ~ 40 -50 MeV. As a corollary, the deuteron has no bound excited states [38,39]. Since each nucleon has isospin $+1/2$, the deuteron has total isospin of either 0 or 1, and so there are four possible triplet and singlet states [40]. The deuteron ground nuclear state is the triplet 3S_1 state with the nuclear orbital angular momentum = 0, and the nuclear spins parallel. States with any nuclear orbital angular momentum of >0 show no signs of binding. The actual structure, and energy, of the deuteron ground state results from its three form factors (electric monopole {charge}, magnetic dipole, and electric quadrupole moments). The magnetic moment of the deuteron ($\mu_D = 0.8574$) is not exactly the sum of its component parts (μ_H and μ_N), and the slight difference -- and the unusual electric quadrupole moment ($Q = 0.282$ e-fm²) -- are both consistent with a more complex mixture of nuclear spin states in the deuteron ground state (suggested by Rarita and Schwinger) such as inclusion of some 3D_1 state [41,42]. There is a virtual (1S_0) state located above the zero binding energy at +30 keV which has been confirmed by nuclear photoeffects and other phenomena [43].

EXPERIMENTAL - ABSENCE OF METACHRONOUS BETA EMISSION

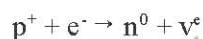
Nickel cathodes produce excess enthalpy (heat) during electrolysis of light water solutions [27]. As a result of competing reactions [44-46], an optimum "notch" occurs in the power gain curve relative to input power [27]. Nickel electrodes were driven within their π -notch using ordinary water in combination with either platinum or gold anodes. We examined several electrodes that had demonstrated episodes of excess heat for their metachronous electron emission, at 12 to 24 weeks, using a modified Maestro-EEG system with solid state detector. Fig. 2 shows the control, the background, and the background subtracted from the sample's emission spectrum. There was apparently insignificant metachronous electron or ionizing radiation emission from the previously-driven nickel cathodes to herald an explanation for the excess enthalpy. The absence of an obvious emission signature to declare ash might be consistent with a short-lived product, or with inadvertent removal of the isotope from the surface during the interval between driving and examination of the samples, or with unintentional self-absorption of the emission signal, or with loss of the signal in the noise background, or even with an unconsidered pathway yielding a stable final product. Given the recent results of Lin [47] and Bockris, and Mizuno, it may be that the delay of several months prior to the metachronous measurement was far too long

and that emissions may have been in the range of 15-25 keV. Studies are underway to reexamine this experimentally. Nonetheless, we considered the latter possibility.

INTERPRETATION -- POSSIBLE PATHWAY

When the possible pathway mentioned above is considered, it is noted that it might contribute to the excess heat and to other researchers' reported transmutation reactions, through the production of *de novo* deuterons. We suggest a multistep pathway leading to deuteron production with the generation of an excited intermediate state (a virtual deuteron in light water with nickel, and -- as presented elsewhere -- an excited helium-4 nucleus in heavy water with palladium systems). Fig. 3 shows some of the relevant nuclear states.

The first step is the corollary of well-known neutron decomposition. Free neutrons have a theoretical lifetime of ~10 minutes but are observed to decay within circa three minutes via interaction with a proton. Taking the neutron lifetime as 918 seconds, we rewrite the basic beta decay equation, using backwards time, to give a proton and electron opportunity to react to an extremely rare occurrence in a fully loaded, one kilomolar electron-loaded, fully-hydrided alloy. This gives a thermal neutron and neutrino.



Could there actually be sufficient quantities of such putative ephemeral thermal neutrons available? Of the estimated 5×10^{21} nickel atoms in the cathode, perhaps the relatively active portion may extend to a depth yielding ~0.04% of the nickel lattice sites which actually are able to have a role and contribute. This constitutes about ~4 micromoles of nickel contributing lattice sites within the cathode. Thus, assuming full local hydridation, the amount of protons available is also in the range of 2×10^{18} . Given the neutron lifetime, there is a statistical likelihood of only 0.000987 contributing at any point in each second. With the putative active sites available, this would give $\sim 2 \times 10^{15}$ potential virtual neutrons per second. This does exceed the required event rate described below.

The second step, similar to the Phuson mechanism proposed for deuteron-laden palladium, involves a hypothesized intermediate precursor-product excited state. Here, the critical excited (and virtual) nuclear state (D^*) is energetically located above the non-bonding axis. The ground-state deuteron forms through a simultaneous cooperative reaction involving a cluster of phonons (the Phuson) linking the de-excitation of the excited virtual deuteron state. These phonons are critical because they account for the coupling, for the focusing of energy into the critical sites, and possibly even for the positive feedback that permits the reactions.

The D^* deexcitation enables the vicinal phonon ensemble to compete effectively and permit fusion reactions to proceed in the fully loaded metals with energy transfer to the phonon cloud. If such nuclear-phonon cloud pathway was not available then the tiny population of D^* would saturate and end all such reactions. Only in the fully loaded material are the virtual levels continuously drained leading to their replenishment. When nuclear-phonon cloud pathways are not available then the tiny population of D^* immediately saturates to an insignificant level.

Our calculations suggest that in order to conserve momentum and energy not all the ~2.2 MeV binding energy would be available. The four-vector relates the momenta (p_i ; $i = x, y, z$) and energy (E), and begins in the center of mass frame-of-reference at the primary reaction site within the hydrided metal. Although several approximations have been made, the energy of the reaction actually available to the Phuson cluster appears to be closer to 1.4 MeV per transition; somewhat less than the binding energy. Because each watt requires $\sim 6.24 \times 10^{12}$ MeV per second, one impact of this correction is that this putative pathway would require $2.8-4.5 \times 10^{12}$ events per second (Fig. 4).

Assuming a distribution of phonon energies in the range of 30-50 eV, there would be $2.8-7.3 \times 10^7$ phonons in the phuson cluster (the cooperative phonon group linked to the excited state). Thus, this would require the involvement of about 5-44 million lattice sites, and therefore about $\sim 170-350$ lattice sites in any direction radial from the prime reaction site. Now, any putative deuteron production releases energy on the $\sim 1-2$ MeV scale at a single location, but with a possibility for spreading the released energy among these lattice sites that can be coupled during the deexcitation time of D^* . The interaction radius $[r_{DE}]$ is determined by the speed of light, c , so the number of lattice cells involved in a single event, coupled through the Phuson deexcitation pathway, assuming isotropicity, is

$$N_{sites}(r_{DE}) = \frac{[c * \tau_{DE}]^3}{[V_{unitcell}]}$$

The uncertainty principle can estimate the lifetime based upon the "energy bandwidth" of the energy involved. For the excited state of deuterium located at +30 keV, the energy width of the virtual state must be on the order of a fraction of that, which would be consistent with an adequate lifetime of the excited state. The ensemble of phonons, with consideration of the available lattice sites, is thus sufficient to account for energy transfer to the lattice during the deexcitation D^* .

LIMITS OF DEUTERON DETECTION

Several experiments are suggested based upon the Phuson theory. First, if deuterium is an ash, and if there are no secondary reactions (e.g., to convert the generated deuterium to helium-4), then there will be an ultimate limit to the amount of energy which can be obtained by loading nickel with a fixed volume of light water. Per mole of hydrogen converted (in ~ 9 cubic centimeters of ordinary water) this would theoretically generate a maximum energy of $\sim 10^{11}$ joules. Second, if the putative thermal neutron production does occur, then there is a maximum excess rate of heat production available based upon that factor, too. This would limit the rate of generation of the desired product. The observed π -notches could be consistent with this.

Third, the rate of generation of *de novo* deuterium per megajoule, if any, is miniscule and would create only a few percentage change in a background of ordinary water (Fig. 4). Such small production levels, and the inevitable separation factors (Fig. 1) refining further possibly already present deuterium, together pose significant challenges for the unambiguous identification of deuterium. Reexamination of vibrational spectroscopic data [48] (Fig. 5) indicates that small amounts of deuteron production remain below the present selectivity and sensitivity of the system. However, other spectroscopies that may offer potential for *in situ* examination include infrared absorption analysis, neutron reflectometry and, nuclear resonance broadening (NRB) and elastic-recoil-detection-analytic (ERDA) techniques. Some techniques penetrate subsurface structures to examine the magnetic properties of, and the impact of nonequivalent occupied sites within, the hydrided metal.

Fourth, if Phusons are involved, then given the requirement for sufficient lattice recruitment, any observed isotopic changes in transmutation systems may occur in greatest amounts below the lattice surface, and at depths which are the order of the interaction radius. Fifth, if the putative thermal neutron pathway exists, then there may also be the potential for nickel (and other elements) to be transmuted to higher isotopes of the same element with the unstable isotopes becoming, for the nickel, the stable isotopes of copper and cobalt.

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37. This heavy water distillation machinery for commercial deuterium production involves towers of 35 meters height with a 9 meter diameter operating in countercurrent exchange mode. These towers must perform at high pressure in anticipated severely corrosive environments (> 6 mm allowance). In the GS system, hot hydrogen sulfide ($70\% \text{H}_2\text{S}$) rises through falling water in a series of cascades; providing enrichments of $\sim 30\text{-}40\%$. The GS system uses final enrichment of electrolytic means so as to produce 99.75-99.8% deuterium oxide (reactor grade heavy water). The ammonia hydrogen exchange system requires an additional catalyst. The high temperature is used since the forward reactions toward material deuteration (from the contamination levels in the water) are then favored. These reactors contain toxic, flammable, and corrosive substances. (Cf. H.K. Rae, "Separation of Hydrogen Isotopes," ACS Symposium, Series 68, Washington D.C., 1978.)
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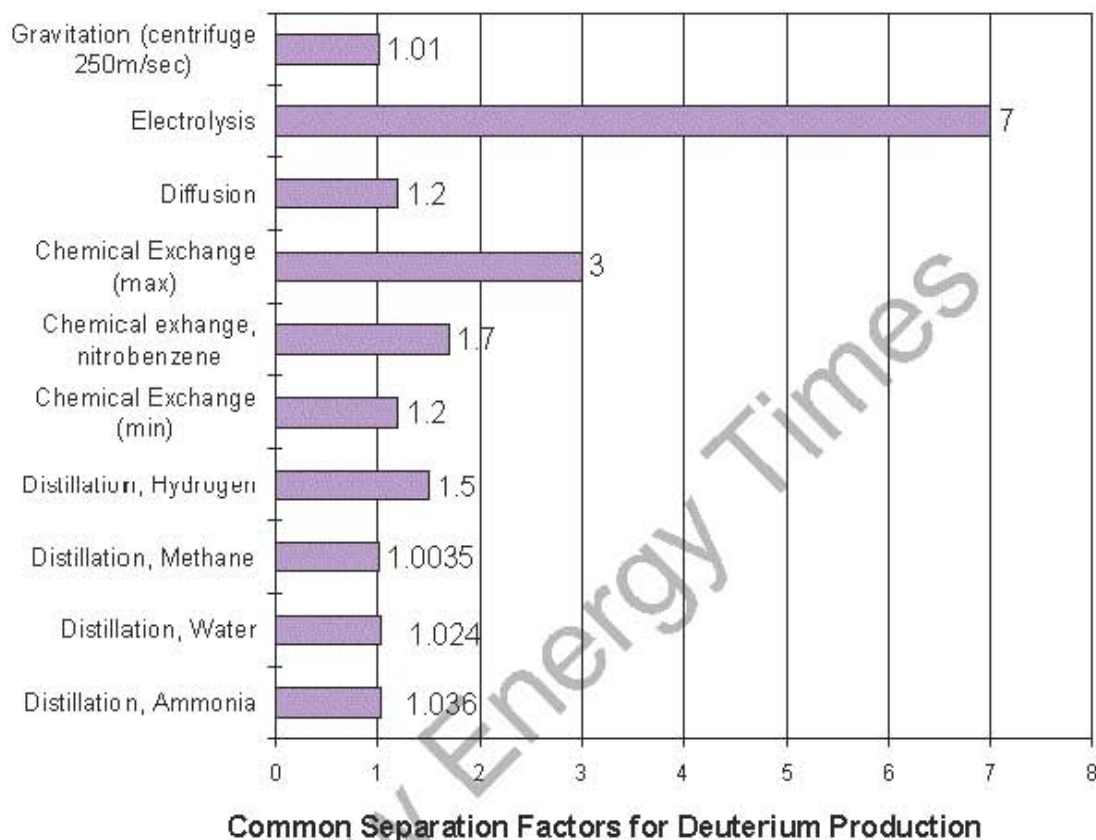


Fig. 1. Common Separation Factors for Deuterium Production

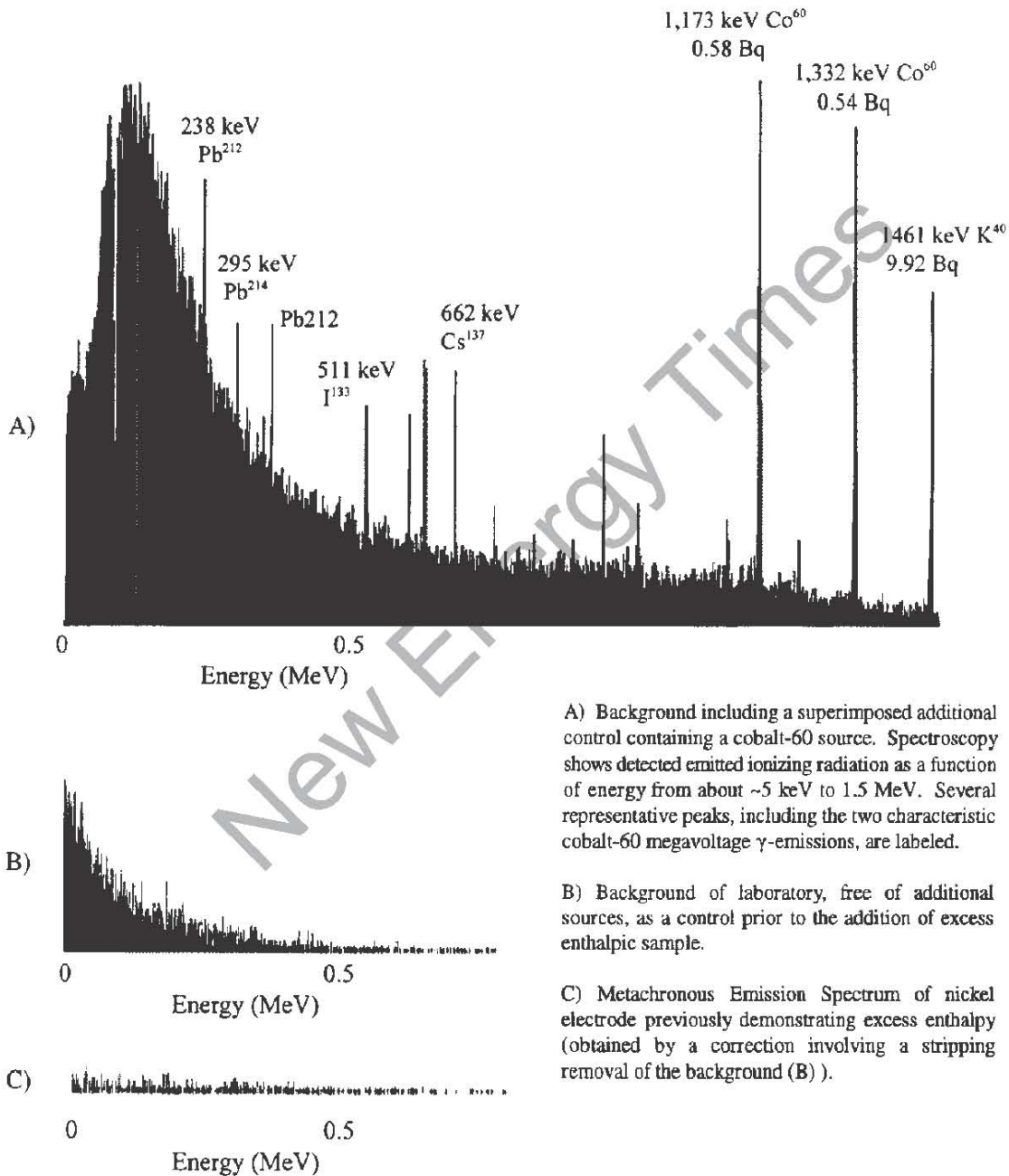
This Figure shows the separation factors for refining deuterium from ordinary water. The values range from a maximum for electrolysis to those values characteristic of evaporation or centrifugation.

Table I

HYDROGEN ISOTOPE PROPERTIES (ref. 49)			
Baryon/Nucleon	Proton	Deuteron	Triton
Mass (amu)	1.0078	2.0142	3.0160
Electric monopole charge	1+	1+	1+
Magnetic dipole moment	2.7928	0.8574	2.9789
Electric quadrupole moment (nuclear magnetons)	0	0.282 e-fm ²	0
Isospin (parity) J	1/2+	1+	1/2+
Binding energy	0	2.224 MeV	6.461 MeV
Lifetime	stable	stable, fragile	12.3y
Decomposition product	not applicable	not applicable	18.59 keV beta
Diatomic gas	Hydrogen	Deuterium	Tritium
Molar volume of solid cm ³	28.3	23.5	
Triple Point (deg K)	13.92 K - 13.96 K	18.58 K - 18.72 K	20.6 K
Heat of vaporization at triple point Joules/mole	910.9	1268.2	
Melting point (deg K)	13.95	18.65	
Boiling point (deg K)	20.38 - 20.4	23.5	
Latent heat of fusion (melt) Joules/mole	117.2	197 - 217	
Heat of dissociation KJoules/mole	437.5	439.2	
Water (ignores HDO, HTO, etc.)	H ₂ O	D ₂ O	T ₂ O
Background concentration	1	0.000156	1/10 ¹⁷
Molecular weight (C ¹² scale)	18.015	20.028	
Surface tension (20C)	72.75 dynes/cm	67.8 dynes/cm	
Temperature of Maximum density	3.98 C	11.23 - 11.6 C	
Dielectric constant (25 C)	78.54	78.25	
Absolute viscosity (20 C)	10.09 millipoises	12.6 millipoises	
Melting point (deg C)	0	3.82	
Boiling point (deg C)	100.00	101.42	
Specific gravity (20 deg C)	0.9982	1.106	
Heat of fusion Joules/mole	6008.2	6317.8	
Heat of vaporization Joules/mole	43,840	44,936	
Solubility NaCl (25 C) gms/100 g	35.9	30.9	
Autopyrolysis constant [D ₃ O+] {OH-} 25 C	1.00 10 ⁻¹⁴	3.00 10 ⁻¹⁵	
pH	7.00	7.26	
Cost	rain, snow	1\$/L(gas) \$100 - \$1000/L (liq)	~ \$30 K/gm
World production	-	~ 1-3 10 ⁶ L/year	< 10 ² Ci/yr
World capacity	-	10 ¹³ tons on surface	

Fig. 3. Metachronous Emission of Nickel Cathodes

This Figure shows Emission Spectrograms of a control, of the background, and of a nickel cathode which had previously demonstrated excess heat (~52 kilojoules, 0.35 cm³, 28 cm², 15 weeks prior to examination).



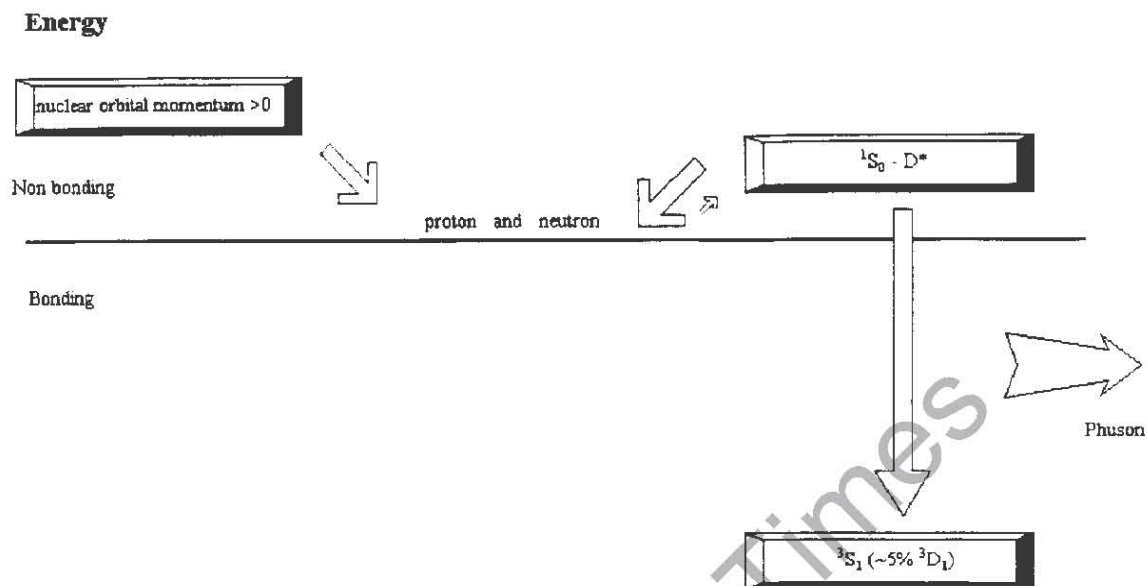


Fig. 3. Spin Manifolds of the Deuteron

This Figure shows the pathway leading to possible deuterium production. It does not include the preceding generation of the neutron as discussed in the text. Shown are three of the "states" available to an interacting proton and neutron. The vertical axis is energy and is not quantitative. If there is any nuclear orbital angular momentum then the non-binding "state" on the upper left is not achieved for reasons discussed in the text. If the nuclear orbital angular momentum is zero, the manifold on the right can be entered. The singlet state above the axis (~ 30 keV) is a virtual state observed in incident beam studies. The state on the bottom right is the well-known ground state of the deuteron, which is mainly a 3S_1 state. The hypothesis is that there may be leakage from the virtual state to the ground state with Phuson coupling to the solid hydrided lattice cathodically loaded with electrons.

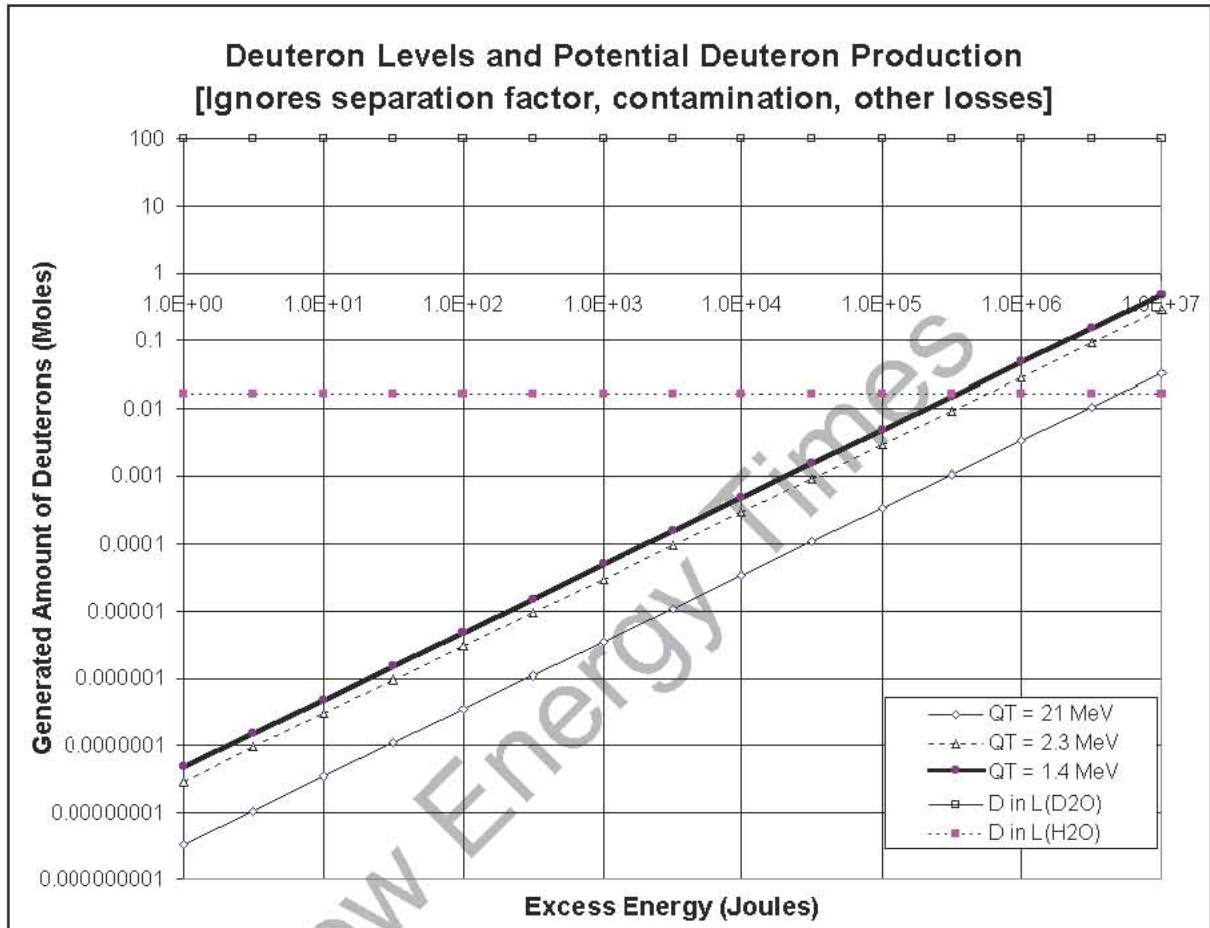


Fig. 4 Putative Production Rates of Deuterium Would Present Detection Challenges

This Figure shows the quantity of deuterium present in, or generated *de novo* in, water as a function of excess energy. The three rising curves show the amount of expected deuteron ash which would be produced if that reaction did occur and did only account for the observed excess heat. Each curve represents a different derived energy for each single event nuclear reaction. The event (binding) energy of 21 MeV does not hold for deuterium, but is representative of what occurs for other final states such as helium. Q_T of 2.3 is what might be expected for the binding energy of the deuteron. The curve representing " Q_T " of 1.4 (thick diagonal line) is derived from the calculations which correct for momentum transfer to the Phuson cluster (see text). Also shown for comparison is the quantity of deuterons contained in a liter of light, and heavy, water.

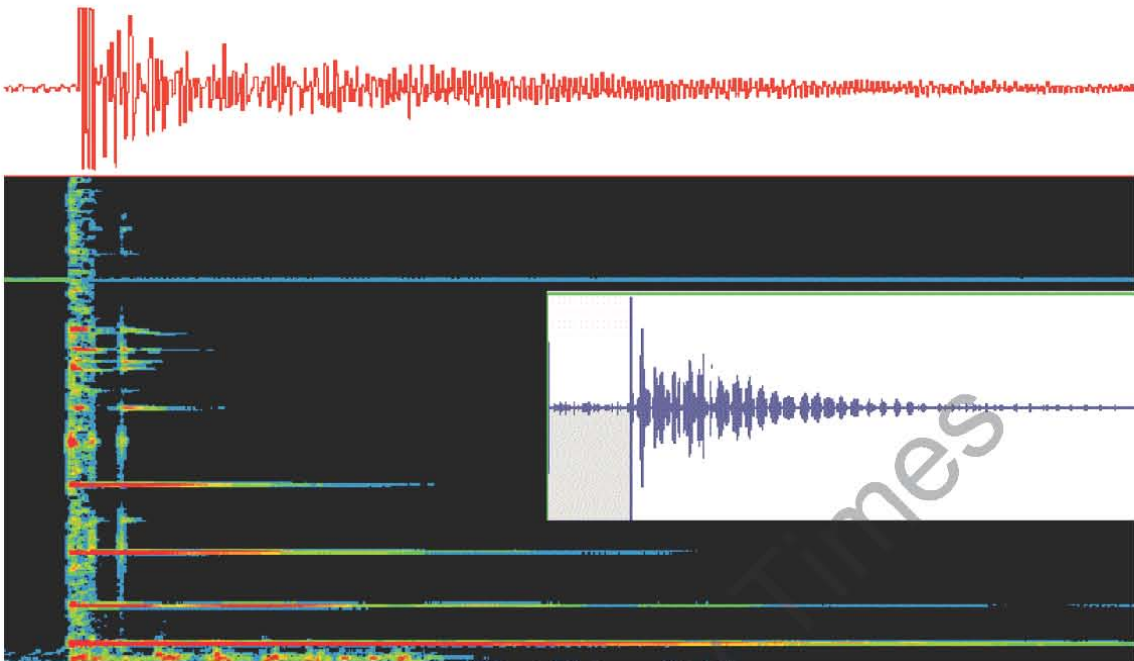


Fig. 5. Vibration Spectroscopy of Nickel Cathode

This figure shows a Vibration Spectrogram of a loaded nickel cathode. The cathode (Sample 92-505b/Ni-B2) surrounded with 20 ml of ordinary water, during its electrical polarization with a platinum anode, is struck with a single mechanical pulse at $t = 0$. The vibrational modes of the electrode, and their damping, can be used to determine loading, but because of separation issues may be insensitive to any putative deuteron production. Two recorders were used to pickup the data (top and inset), and the fast fourier transform is also shown as a frequency versus time plot. The solid bar three-quarters of the way up the calibration signal at $17,390^{+/-53}$ Hz. The lowest evanescent normal mode, the most irregular of the group, is believed to be a circular mode around the relatively large annular electrode. The sampling rate was ~ 100 kHz.

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