COMMENT

COLD FUSION PRODUCES MORE TRITIUM THAN NEUTRONS

FLEISCHMANN *et al.*[1] reported the occurrence of nuclear fusion during the electrolysis of 99.5% D_2O (containing 0.1 M LiOD) using a palladium cathode. They found emission of neutrons, accumulation of tritium in the electrolyte (as a result of electrolysis), and generation of excess heat over and above that due to the following electrode reactions:

cathode reactions.

$$D_2O + e \rightarrow D_{ads} + OD^- \quad (1)$$

$$D_{ads} + D_2O + e \rightarrow D_2 + OD^- \quad (2)$$

$$D_{ads} + D_{ads} \rightarrow D_2 \quad (3)$$

balancing anodic reaction

$$4\,OD^- \rightarrow D_2O + O_2 + 4\,e. \quad (4)$$

The net heat due to the reactions (1), (2), (3) and (4) is zero, and these reactions contribute a cell voltage of 1.54 V, which has been referred to, by these authors, as the voltage for the above thermoneutral reactions. In addition they also observed the emission of γ-rays from the electrolytic cell. To explain these observations, they proposed the occurrence of the following nuclear (fusion) reactions in the Pd cathode:

$$^2D + {}^2D \rightarrow {}^3T\,(1.01\ MeV) + {}^1H\,(3.02\ MeV) \quad (5)$$

$$^2D + {}^2D \rightarrow {}^3He\,(0.82\ MeV) + n\,(2.45\ MeV). \quad (6)$$

The generation of γ-rays was explained to be due to the (n, γ) reaction

$$^1H + n\,(2.45\ MeV) \rightarrow {}^2D + \gamma\,(2.5\ MeV). \quad (7)$$

At about the same time Jones *et al.*[2] reported (based on their observation of neutron emission) cold fusion in the electrolysis of D_2O (99.5%) containing a mixture of salts, with a titanium cathode.

These reports have created great stir among various research groups all over the world. As a result, there have been many confirmations and denials. However, only a few of the confirmations have been published in scientific journals. Reports of failure to duplicate the results of Fleischmann *et al.* have been discussed in the special meetings organized to understand cold fusion but most of them have not been published. Mathews *et al.*[3] found a significant amount of neutron emission over the background in the electrolysis of D_2O (containing nickel and palladium chloride) with Ti cathode. They also reported that the cathode was hotter in D_2O electrolysis compared to H_2O electrolysis. Santhanam *et al.*[4] reported production of excess heat (over and above that of the electrode reactions) in the electrolysis of 1 M NaCl in D_2O. Electrolysis using a Pd cathode yielded greater excess heat compared to electrolysis using a Ti cathode. γ-Rays were also detected during electrolysis. The radiation level was 48% higher than the background in one experiment while it was only 8% more than the background in another experiment. The Central Electrochemical Research Institute, Karaikudi, came up with a confirmation of cold fusion[5] based on calorimetric measurement and detection of tritium (by autoradiography) embedded in the Pd cathode.

Confirmation of cold fusion was reported by Scaramuzzi at the workshop on cold fusion held at Santa Fe (May 23–25)[6]. This was a different type of experiment, carried out at Frascatti. The logic for doing this experiment appeared to be that, in the electrolysis experiment, cold fusion was the result of interaction of D atoms in the metal lattice, and the role of electrolysis was to load the metal with D_2. In the light of this argument they loaded Ti pieces with D_2 gas from a D_2 gas bottle at 8 atm pressure and liquid nitrogen temperature. When the D_2-loaded Ti was heated to 200°C, with simultaneous evacuation, there was emission of neutrons. Also reported was the work of Magni of the ENFA CASSACCIA, who used a sponge of Ti loaded with D_2, which was cooled and then pumped. A small peak of neutrons was observed after about 700 min. Bursts of neutrons were also observed by Massoni *et al.* when blades of Ti (0.5 mm thick) were heated quickly (20 s) to 1000°C and cooled in gaseous D_2 to 500°C.

Recent results of several research groups of the Bhabha Atomic Research Centre (BARC), Bombay, reported by Dr P. K. Iyengar[7], reveal new and interesting features of cold fusion. Electrolytically induced cold fusion has been investigated by BARC employing a Milton-Roy cell (a commercial cell used for production of H_2) with Pd-Ag tubular cathode, and cells made in the laboratory with Pd cathode. They monitored both production of neutrons and accumulation of tritium. Both BF_3 detector embedded in paraffin and proton recoil fast

neutron detector showed substantial neutron emission. In the case of electrolysis using the Milton–Roy cell, the BF_3 counter measured 17,758 counts/300 s (background 65 counts/300 s) and the proton recoil neutron detector gave a count rate of 25,872 counts/300 s (background 650 counts/300 s). Taking into account the efficiency of the counters, the neutron emission rate was estimated to be $\sim 2 \times 10^7$ neutrons/300 s. Neutron emission was not continuous but occurred in bursts, and the duration of the 'flashes' was probably under 160 μs. The emission was basically Poisson in nature (i.e. the neutrons were emitted one at a time).

BARC, unlike many other groups, carefully monitored tritium levels in electrolytes, taking precautions to use ^{40}K-free vials to minimize background counts and minimizing chemiluminescence interference effects. Excess tritium produced by electrolysis was computed, taking into account initial concentration of tritium and dilution effects due to periodic make-up of D_2O. Their estimates should be regarded as conservative because tritium locked in the cathode and tritium lost in electrolysis were neglected (it is known that the T/D ratio in gas evolved from the cell is nearly the same as that in the electrolyte). Typical results for tritium enrichment are shown in table 1. On the basis of a comparison between the neutron and tritium production rates, it is reported[7] that cold fusion may be characterized as being essentially 'aneutronic', with a neutron-to-tritium channel branching ratio of less than 10^{-8}. This is a surprising result when viewed in the light of the fact that muon-catalysed D–D fusion has a channel branching ratio of nearly unity (T and n production probabilities are 0.42 and 0.58 respectively[8]). Viewing the occasional burst of 100 neutrons in a single burst in the light of the branching ratio estimate of 10^{-8}, the report[7] states an intriguing conclusion, viz. that a chain reaction involving as many as 10^{10} fusion reactions must be occurring within 160 μs.

BARC reproduced with success[7] the Frascatti experiment. Ti pieces were exposed to D_2 gas at 50 atm. When this charge was subjected to temperature cycling between 77 K and 300 K emission of neutrons was noticed. It was not constant but exhibited several peaks. Typical count rate for a peak was 3900/40 s (background 60/40 s). In a 7 h duration the neutron emission was $\sim 6.5 \times 10^6$ neutrons. In another experiment D_2 gas was loaded into small Ti machined targets (discs, cones, etc.) at 1 atm pressure and 900°C (temperature attained by induction heating). Switching off the induction heating cooled the charge, and Ti absorbed D_2 gas in a minute. The targets showed statistically significant excess neutrons (over the background) during the gas absorption. One of the discs emitted neutrons nearly 50 h after loading and produced in all $\sim 10^6$ neutrons during an 85 min active phase.

Perhaps the most significant experiment carried out by BARC was that of establishing the presence of tritium in Ti loaded with D_2 gas. If D–D fusion took place in gas-loaded Ti, atoms of tritium should be produced and they must be locked up in the Ti lattice. Autoradiography of deuterated Ti discs showed randomly distributed intense spots, indicating that tritium was not uniformly distributed but was segregated in small regions that were random in location.

Another experiment for confirming the presence of tritium in gas-loaded Ti is an even more elegant one. Tritium is a β emitter with a half-life of 12.4 years. The end-point energy of betas is 18 keV. It is to be expected that β from tritium locked in Ti should excite K_α and K_β X-rays of Ti since their energies are, respectively, 4.51 keV and 4.931 keV. Using a Si (Li) detector, they found both K_α and K_β of Ti from D_2-loaded Ti samples.

I would like to make some comments on the results quoted above. There has been considerable criticism of the excess enthalpy data reported by Fleischmann *et al.* Some of the criticism seems to be

Table 1 *Production of tritium during electrolysis of D_2O*

Cathode	Electrolyte	Volume of solution (ml)	Tritium levels (Bq/ml) initial	final (10^3)	Tritium produced (no. of atoms) (10^{14})
Pd–Ag tube (M–R cell)	5 M NaOD in D_2O	250	2.6	55.6	80
Pd sheets	"	1000	2.0	7.0	40
Ti rod	"	135	2.0	1.8	1.3
Pd cylinder	0.1 M LiOD in D_2O	65	18.1	8.8	3.0

due to some misunderstanding of the term excess enthalpy. By excess heat the authors mean the heat generated over and above that due to the reactions (i) to (iv) and the Joule heat (=cell current × (cell voltage − 1.54)).

Fleischmann *et al.* estimated the heat produced from the nuclear reactions (v) and (vi) using their data of rate of neutron emission ($4 \times 10^4\ s^{-1}$) assuming a channel branching ratio of 1. They found the estimated heat to be much smaller than the experimentally observed heat. On the basis of excess heat observed they expect the fusion rate to be in the range of 10^{11}–10^{14}. In order to explain the tremendous disparity between the fusion rate based on neutron emission rate and that based on calorimetry, they proposed that some other nuclear reaction, which does not produce neutrons but produces only heat, must be occurring. In the light of the recent results of BARC, I feel that this disparity can be understood without invoking some hitherto unknown reaction. The argument of Fleischmann *et al.* was based on equal probability for the occurrence of neutron- and tritium-producing reactions. It is now known that the channel branching ratio for D–D fusion in metal lattice favours tritium production 10^8 times more than neutron production. Therefore a neutron production rate of $4 \times 10^4\ s^{-1}$ (ref. 1) would mean production of 4×10^{12} atoms of tritium per second. Using this figure we calculate the heat expected to be released in the electrolysis of D_2O with a 10-cm-long Pd cylinder of 0.4 cm diameter to be 2.056 W/cm^2, which is approximately 50% greater than the 1.39 W/cm^2 reported by Fleischmann *et al.* This discrepancy is very much smaller than that arrived at on the basis of unity channel branching ratio, and may be explained in terms of heat taken away by the gases, fluctuation in rate of fusion reaction, etc. An excess heat of 1.39 W/cm^2 would be accounted for by production of 2×10^{12} atoms of tritium per second. Thus the tremendous disparity in the fusion rates estimated from the heat generated and the neutron flux observed can be resolved by this explanation without invoking a hitherto unknown nuclear reaction or proposing the following reaction[3]:

$$D + D \rightarrow {}^4He + \text{energy absorbed by the lattice.} \quad (8)$$

This reaction is far-fetched as the nuclear reaction times are several orders of magnitude smaller than the time taken for the lattice to absorb the energy[6]. If this explanation is accepted one has to find an explanation for the estimate of 1–$2 \times 10^4\ s^{-1}$ (for tritium production) arrived at by Fleischmann *et al.* from the measured tritium accumulation and the D/T separation factor. In other words, there is a discrepancy between the BARC results and those of Fleischmann *et al.* in respect of channel branching ratio. The former place it at $1:10^8$ whereas the latter put it at 1:1.

Most of the efforts of research groups appear to have been in verifying the calorimetry and neutron production rate. Confirmation of tritium production has received far less attention, and identification of He in the electrode, the least. Besides those mentioned above Storms (LANL) reported a 100-fold tritium enhancement[9] and Appleby found a million-fold tritium enhancement in 10 h[6]. It could perhaps be more meaningful to look at rate of tritium production.

Comparison of the reported results has become difficult essentially because the papers do not provide data on electrode size, time of electrolysis, number of coulombs passed, etc. For instance neutron production per unit volume of electrode can only be calculated for the results of Fleischmann *et al.* and those of Jones *et al.* The former yield 3.19×10^4 neutrons $s^{-1}\ cm^{-3}$ while the latter give 6.15×10^{-1} neutrons $s^{-1}\ cm^{-3}$. From other reports[3,7] such data could not be calculated owing to lack of details.

To understand cold fusion, solid-state effects have been considered[8, 10–13]. The possibility of increasing the penetrability of the Coulomb barrier by increasing the effective mass of electrons has been discussed. An effective mass of 6.2 would be able to account for the rate based on the neutron emission rate of Fleischmann *et al.*, and an effective mass of 11.4 would account for the rate derived from the heat data of these authors[11]. However, such a solid-state effect is not considered feasible since the effective mass effect is meaningful only for electronic motion over lengths greater than that of interatomic spacing and is irrelevant for movement over the much smaller distances involved in fusion reactions. Taking a cue from the observation that neutrons are emitted by an LiD crystal when it is fractured[14] and the suggestion[15] that cracks may be the site of fusion reactions, Kumar[13] suggests that a propagating crack in embrittled Pd or Ti may result in acceleration of deuterons to kinetic energy $\sim 10^2$–10^3 eV above the equipartition value, and that these deuterons may be responsible for fusion.

Iyengar[7] suggests that the very high probability for the tritium branch in cold D–D fusion reactions is indicative of transfer of neutrons across the potential barrier as suggested by Oppenheimer and

Phillips[16] and elaborated recently by Rand McNally[17]. To account for the deuteron-induced radioactivity*[18] Oppenheimer and Phillips[16] gave the following explanation. Reactions of this type can occur without requiring the penetration of a charged particle through the nuclear Coulomb barrier. When a deuteron is projected against a nucleus, the proton is held back by the Coulomb field. The neutron, which is rather weakly bound, is immune to the action of the electrostatic field, and its probability distribution can overlap the centre of the nucleus. This distribution is in fact distorted by the forces involved in the deceleration of the neutron, in such a way as to increase its density on the side towards the nucleus. Thus the neutron, which is loosely bound (binding energy ~ 2 MeV) in the D nucleus, is transferred to the target nucleus.

In the light of these suggestions we may propose the following hypothetical picture for the cold D–D fusion. When a metal like Pd or Ti is loaded with D_2, crack formation may occur owing to volume change. Propagation of such a crack could accelerate deuterons. (The cracking sound reported by Santhanam *et al.*[4] is indicative of such crack propagation.) These deuterons may bombard PdD_2/D held by Pd or Ti, leading to neutron capture and formation of tritium with the release of protons and energy. The released proton might transfer its energy to some other deuteron and thereby start a chain reaction, which would be terminated when a substantial portion of D in the crack tip is transmuted. The reaction would start again when another crack forms. This picture qualitatively explains the occurrence of fusion reactions in bursts and also the random distribution of reaction sites. The statistical nature of nucleation and growth of cracks and their occurrence at any point in the bulk may explain the volume effect noticed by Fleischmann *et al.* as well as the low reproducibility. This picture however fails to explain emission of neutrons. These probably form from a few of the high-energy deuterons generated by a growing crack.

I would like to point out another possibility. The deuterons accelerated by a propagating crack may hit a Pd/Ti nucleus instead of a deuterium nucleus and may transmute Pd/Ti. Such a reaction can be expected to be exothermic by analogy with other transmutations brought about by deuterium bombardment. Since the variation of transmutation yield with atomic number of target is rather small, such a reaction might be possible with nuclei heavier than copper[18]. If this type of reaction occurs, then we would have generation of heat without production of He or tritium. This possibility is worth examining.

*When nuclei such as Na, Al, Si and Cu are bombarded with deuterons (energy 0–3.6 MeV), they capture a neutron and get transmuted into an isotope with mass one unit higher and exhibit radioactivity.

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