

COLD FUSION ONE YEAR AFTER

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ABSTRACT

The one year long history of cold fusion is critically reviewed on the basis of the more recent results, in an attempt to establish the perspectives of this field.

1. INTRODUCTION

On 23rd March 1989 Prof. Fleischman and Dr. Pons announced, in a press conference, the discover of a method to let nuclear reactions happened inside an electrochemical cell at a rate sufficiently high to produce significant amount of heat, suggesting that the energy problem of the world would have been definitely solved in few years. The social relevance of this matter, the economical and scientific interests, the way chosen to announce the discovery making very heavy use of the media, started one of the biggest scientific dispute science has ever been confronted with, the echoes of which still are present, and that appreciably modified the perception society has of science.

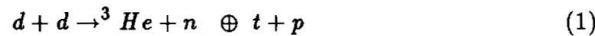
In this lecture we will discuss of the status of the cold fusion searches one year after the claimed discovery. Many groups have attempted to reproduce the original results, many of them were unable to found any significant evidence of nuclear reactions or anomalous heat production, some found unexpected and mostly irreproducible production of heat or of nuclear products. In any case a lot of work and research has been performed and it is instructive to try to establish which is the situation now and to guess which are the chances that nuclear fusion could take place at a detectable rate in deuterated metals at room temperature.

A basic rule of a scientific discussion, is to report facts in a objective way, for instance giving numbers and detailed description of the experiments performed, and we will try to stick to this rule. However given the large use of mass media, email and faxes to circulate private communications on the subject, some of the informations reported here, while being relevant for the discussion, are not taken from scientific publications. In addition, given the heat that developed around this

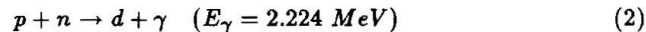
subject, it is possible that some of our statements look biased and we apologize in advance. However, during the time we spent working on cold fusion we have been often faced with many unexpected experimental results and learned how it was rather difficult to accept all of them at the same time. We were indeed forced by this very fact to the attitude well described by the words of D. Hume¹, who wrote (approximately): "Would you rather that all the laws of Nature be violated or believe that one man made a mistake?". We are convinced it is likely (in few cases out of question), that in the CF business, some people made mistakes and sometimes repeatedly.

2. THE THREE DISCOVERIES OF CF.

Cold Fusion mania begun with the noisy announcement given to the media by the University of Utah, that Prof. Martin Fleischman (Department of Chemistry of the Southampton University, Great Britain) and Dr. Stanley Pons (University of Utah, United States) (FP in the following) observed large excess of heat, operating electrochemical cells with heavy water solutions. In a paper titled *Electrochemically induced nuclear fusion of deuterium*, initially faxed around the world and later published on the Journal of Electroanalytic Chemistry², they described experiments performed using electrolytic cells with platinum anodes and palladium cathodes, in a solution 0.1 M of LiOD in 99.5% D₂O + 0.5% H₂O. The Pd cathode was electrolytically preloaded with deuterium. When the equilibrium was reached, the solution temperature was continuously monitored to measure possible excess of heat. Assuming that the observed excess of heat was due to deuterium fusions, through the reactions (from now on $p \equiv {}^1H$, $d \equiv {}^2H$ and $t \equiv {}^3H$):



two other measurements were performed to detect nuclear products. A photon detector was used to search for γ rays emitted in the recombination of the neutrons with protons in the bath surrounding the cell:



and tritium content of the electrolyte was measured repeatedly during the experiment.

The excess of heat reported was large, up to 10 W/cm³ of cathode, as shown in the table taken from the FP paper (Fig. 1). The data were suggesting an effect depending on mass more than on surface: for the three rod shaped electrodes the excess of heat per unit of volume was clearly increasing with the rod size, while in the case of a sheet electrode no excess of heat was seen. In addition the result

Electrode type	Dimensions /cm	Current density /mA cm ⁻²	Excess rate of heating/W	Excess specific rate of heating/W cm ⁻³
Rods	0.1 × 10	8	0.0075	0.095
		64	0.079	1.01
		512 [*]	0.654 [*]	8.33
	0.2 × 10	8	0.036	0.115
		64	0.493	1.57
		512 [*]	3.02 [*]	9.61
	0.4 × 10	8	0.153	0.122
		64	1.751	1.39
		512 [*]	26.8 [*]	21.4
Sheet	0.2 × 8 × 8	0.8	0	0
		1.2	0.027	0.0021
		1.6	0.079	0.0061
Cube	1 × 1 × 1	125	WARNING! IGNITION? See text	
		250		

* Measured on electrodes of length 1.25 cm and rescaled to 10 cm.

Figure 1. Generation of excess enthalpy in Pd cathodes as a function of current density and electrode size (from Ref. 2).

of an experiment with a cubic electrode was not reported, but it was labeled with the somewhat mysterious statement *warning! ignition?*

The paper contained two figures regarding the detection of nuclear products. The first showed the difference between the γ spectrum measured by an NaI detector close to the cell and the spectrum measured in another place of the laboratory near an identical cell not operated. This spectrum showed a clear peak in the region around 2.2 MeV. The second figure showed the spectrum of the scintillating light obtained in the the measurement of the tritium content of the electrolyte; the distribution was peaked at about 3 keV, corresponding to the mean energy of the electrons produced in the tritium β decay. However the tritium contamination of the heavy water used in the experiment was not reported. As the authors pointed out, the amount of nuclear products detected was more than 9 orders of magnitude below what was requested by the theory of the nuclear fusion to explain the observed excess of heat.

Few days later, the "second discovery" of CF. Another Utah group lead by Steven E. Jones (Brigham Young University) (J in the following) circulated a paper also claiming the observation of electrochemically induced CF. J used mostly Ti cathodes and a different electrolyte, made of nine different metallic salts dissolved in heavy water. J stated explicitly that they did not preloaded the cathodes with deuterium; in addition the excess of heat was not measured in these experiments. Instead the energy spectrum of the neutrons emitted was

measured by mean of a rather sophisticated neutron spectrometer developed by the authors. The spectrometer contained a scintillating moderator and a set of ${}^6\text{Li}$ glasses to trigger on thermalized neutrons by the reaction $n + {}^6\text{Li} \rightarrow t + {}^4\text{He}$. The neutron energy was measured using the scintillating light while the neutron signature was given by the coincidence between the light pulses emitted by the scintillator and by the ${}^6\text{Li}$ glasses. The direct measurement of the neutron spectrum is clearly important when searching for deuterium fusion, because the neutrons emitted in reaction (1) have a characteristic energy of 2.45 MeV. Indeed, in one experiment, out of 14 reported, there was a statistically significant deviation from the background in the 2.4 MeV region: in the remaining experiments the effects were smaller, if any. These results were obtained after the subtraction of the background spectrum. The authors obtained from these data a fusion rate of about 10^{-23} s^{-1} (per pair), about 4 orders of magnitude below that reported by FP (from n data).

On 12th April 1989, the first meeting on CF was organized in Erice. After hot discussions, no definite conclusions were reached. However it is instructive to recall some of the comments made at that time³: *...experiments will show whether CF is taking place; if so, it will teach us much besides humility I bet against its confirmation* (R. L. Garwin), *Somebody is going to have to eat his hat* (L. Maiani), *We are also human, and need miracles, and hope they exist* (L. Ponomarev).

Six days later, an Italian group lead by Francesco Scaramuzzi (ENEA Frascati) (in the following S) announced what we will call the "third discovery" of CF. This group abandoning the original approach of the electrochemical cells, performed a set of experiments using Ti shaves exposed to gaseous D_2 , and varying temperature (between liquid nitrogen and room temperature) and pressure (from vacuum to about 50 atm). The fraction of D_2 absorbed by Ti and the temperature were not monitored. They only recorded the rate of (just) one BF_3 neutron counter. The group published a paper⁴ discussing the result of two experiments where the rate, integrated every few minutes, showed a very large increase over background lasting for tens of hours. The first experiment was performed at LN temperature (77 °K) and high D_2 pressure (50 atm) (absorption experiment) while the second was at room temperature and in vacuum (desorption experiment). In the first case the integrated counts show a strange two value quantization, not present in the second case.

After less than a month from the initial announcement three very different experiments, both from the point of view of the techniques used and of the results obtained, claimed to have obtained evidence for CF in deuterated metals at room temperature. These results were suggesting three very different CF scenarios:

- a. FP observed anomalous heat production and a large (but insufficient to explain heat) flux of neutrons of about $2 \cdot 10^4 \text{ g}^{-1} \text{ s}^{-1}$, working in

equilibrium condition with deuterium loaded *Pd* cathode;

- b. J working in non equilibrium conditions with a *Ti* cathode observed a very small neutron flux of about $0.1 \text{ g}^{-1} \text{ s}^{-1}$;
- c. S working in non equilibrium conditions, in the gaseous phase, reported a flux of about $50 \text{ g}^{-1} \text{ s}^{-1}$.

Two of the three experiments used rather poor instrumental techniques (FP, S) and detected very large effects, while the third (J) using a rather sophisticated detector reported a result statistically not too significant. All the three were not redundant in the measurement of nuclear products, in particular neutrons. Both J and S were using a single detector, a dangerous situation when looking for rare neutron bursts in normal laboratory condition. In fact, it is remarkably difficult to implement coincidence techniques in neutron detection. This comment applies particularly to S, where only the number of pulses above a certain threshold were recorded, without any pulse shape analysis. FP detected γ induced by deuterium recombination instead of n , but also in this case the implemented background subtraction is inadequate as we will show in the following.

Since the beginning CF phenomenology was plagued by the problem of irreproducibility. At the beginning of a new field, reproducibility might be difficult to achieve. Soon however, this problem has to be solved otherwise the observed phenomena quickly becomes science or, worse, object related to faith more than science (as clearly has happened with CF).

From a probabilistic point of view if the chances of discovering a large unexpected effect in a well established field like nuclear physics is very low, the probability of discovering two or three different effects seems, at first view, negligibly low. FP showed a huge deficit between observed nuclear products observed and heat excess. If their results were correct one should completely rewrite the theory of nuclear interactions at low energy! Alternatively one should assume that there are large inconsistencies between the different measurements reported.

Before continuing our brief history of CF, we introduce the framework of nuclear fusion and we discuss some aspects regarding the detection of nuclear products, calorimetry and deuterated metals.

3. THEORETICAL AND EXPERIMENTAL BACKGROUND

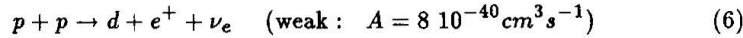
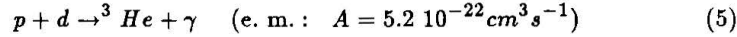
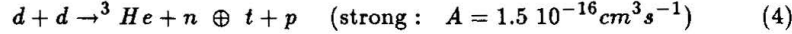
3.1 Theoretical aspects

Cold fusion of light nuclei is not a new branch of physics. From the 50's the so called muon-catalysed fusion, i.e. the fusion of a molecule composed by a meson and two hydrogen isotopes, is a well studied phenomenon ^{5,6}. Other mechanisms, such as the piezonuclear effect ⁷, have been proposed to enhance the fusion rate. After the first claims for CF catalysed by a solid state environment a large amount of theoretical work has been done to understand the experimental results.

Nuclear fusion includes two distinct phenomena: *i*) the nuclear reaction which transforms a couple of light nuclei in a more massive one, and *ii*) the overcoming of the electromagnetic barrier between the charged reactants. This can be seen writing the fusion rate Γ (number of fusions per time unity) between two nuclei as ⁵:

$$\Gamma = A |\psi(0)|^2 \quad (3)$$

where $\psi(R)$ is the wave function for the relative motion of the two reactants (interacting via a spherically symmetric potential) and A a constant depending on the specific fusion reaction considered. Point *i*) concerns the value of the constant A . This changes drastically with the interaction involved in the reaction, i.e. strong, e. m. or weak. For instance, in the case of hydrogen isotopes, we have ^{8,9}:



On the other hand point *ii*) concerns the value of the zero-distance density $|\psi(0)|^2$. This can be expressed ¹⁰ in terms of the density of the nuclei in the medium, $|\psi(\infty)|^2$, by:

$$|\psi(0)|^2 = |\psi(\infty)|^2 \frac{2\pi e^2}{\hbar v} G(E) \quad (7)$$

where $E = \mu v^2/2$ is the relative kinetic energy of the reactant pair with reduced mass μ and e is the electric charge of each nucleus (hydrogen isotopes). The Gamow factor, $G(E)$, depends on the shape of the pair potential:

$$G(E) = \exp\left\{-\frac{2}{\hbar} \int_{R_c}^0 dR \sqrt{2\mu[V(R) - E]}\right\} \quad (8)$$

with $V(R_c) = E$. In vacuum $V(R)$ is the Coulomb potential e^2/R . In a solid state environment the pair potential is generally reduced by screening effects and the fusion rate is enhanced. Nuclear fusion catalyzed by a metallic host is then a quite reasonable concept. Quantitative calculations take, however, some care.

The fusion of two hydrogen nuclei in a solid state environment is a true many-body problem, then approximations must be done to introduce the concept of pair potential appearing in Eqn. 8. The paper by Legget and Baym¹¹ is illuminating in this sense. These authors rigorously prove that any spherically symmetric pair potential which is a lower bound to the fully interacting many-body potential, gives an upper bound to the exact value of the fusion rate when calculating Γ . Using a simple lower bound pair potential Legget and Baym deduce a crude upper bound for the processes in Eqn. 4-6, $10^{-47} s^{-1}$ and $10^{-41} s^{-1}$, respectively. In the spirit of Ref. 11 more severe upper bounds to the fusion rate can be obtained by using more realistic pair potentials.

Before to proceed in this analysis let we introduce some properties of metallic hydrides. We concentrate on hydrogen in palladium¹². *Pd* crystal is a face centered cubic (fcc) lattice. The lattice structure of palladium hydride, *Pd(H_n)*, represents an isotropically expanded form of the fcc host lattice with the hydrogen atoms occupying the so called octahedral sites as shown in Fig. 2. No experimental evidence exists that the hydrogen concentration, n , can be greater than unity. Below about 300 °C the homogeneous solid solution disintegrates into an α phase with low hydrogen content ($n \leq 0.008$) and an expanded, hydrogen-rich β phase ($n \geq 0.607$). Isotherms $p(n)$ (vapour tension) of *Pd(H_n)* at different temperatures are shown in Fig. 3. The lattice constants for pure *Pd* and for the α and β phases are $a = 3.890 \text{ \AA}$, $a(\alpha) = 3.894 \text{ \AA}$ and $a(\beta) = 4.025 \text{ \AA}$, respectively. From these data follows an increase of volume of the host lattice in the $\alpha \rightarrow \beta$ transition. The increase of volume is accompanied by a release of enthalpy $\Delta H \sim 0.2 \text{ eV/hydrogen nucleus}$.

Let us return now to the determination of a realistic potential. The correct strategy is the evaluation of the *total* energy of a pair of hydrogen nuclei embedded in a metallic lattice at a distance R ^{10,13-14}. Each hydrogen nucleus determines a local polarization of the unperturbed electronic charge density of the host metal. Within the adiabatic approximation the total energy of the system can be separated into various contributions¹³: *a*) the electrostatic interaction between the two reactants including their electronic polarization clouds, *b*) the electrostatic interaction of the two reactants with the lattice ions and *c*) the nonelectrostatic interaction (kinetic, exchange and correlation energy) between the polarization clouds. In the case of a *Pd* lattice the three contributions can be calculated as a function of R when the nucleus 1 sits in an octahedral site and the other is free to move along a direction connecting the first particle to an adjacent octahedral site. The resulting spherically symmetric potential represents a lower bound to the true

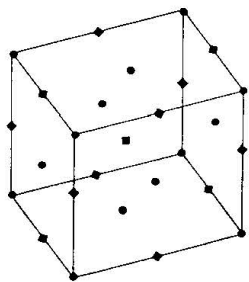


Figure 2. The octahedral sites in a fcc lattice. The black circles represents metal ions and the squares octahedral sites.

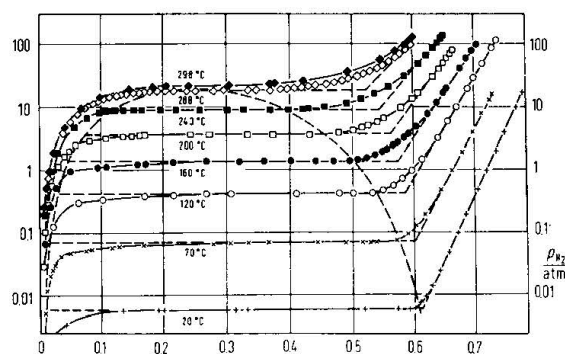


Figure 3. $p(n)$ isotherms of $Pd(H)$ with bulk palladium at different temperatures.

many-body potential (which has no particular symmetry) and the corresponding fusion rate gives an upper limit to Γ .

The depicted pair potential $V(R)$, including the single contributions *a*), *b*) and *c*), is shown in Fig. 4A. The potential has a minimum with a binding energy 1.23 eV . As a consequence hydrogen nuclei pair bound states can exist: the octahedral sites, namely. Depending on the reaction energy two different procedures have to be employed to calculate the fusion rate. In the case of low reaction energy, $E < 1.23 \text{ eV}$, the reactant pair is in a bound state. Then, the relative wave function at zero distance can be calculated by numerical integration of the corresponding Schrödinger equation and the fusion rate is given directly by

Eqn. 3. In the case of high-activation energies, $E > 1.23$ eV, the reaction is *in flight*. For a given hydrogen density in Pd the fusion rate is given by Eqn. 3 and Eqn. 7 via the Gamow factor. The resulting fusion rates for the reactions (2-4) are shown in Fig. 4B as a function of the excitation energy E assuming an hydrogen concentration $n = 1$. It is interesting to note that at low energies the weak reaction (4) becomes more favourite than the strong one (2). In fact the smallness of the fusion constant A is compensated by the growth of the tunneling probability through the electromagnetic barrier due to the lighter reduced mass of the reactants.

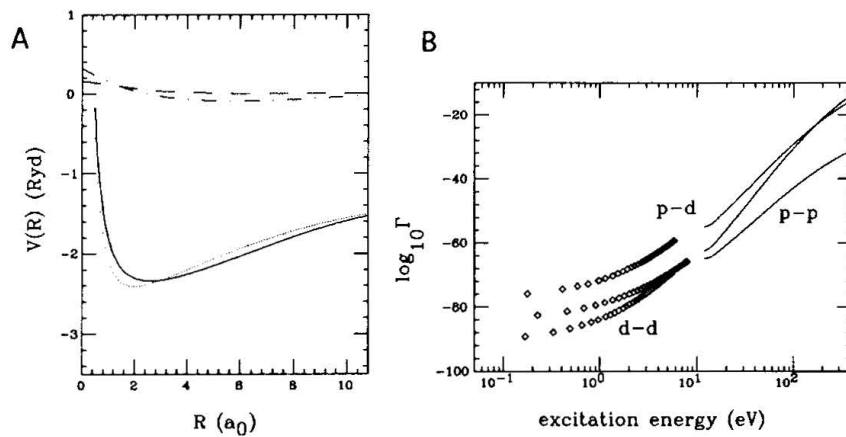


Figure 4. A: Shape of the pair potential $V(R)$ calculated along the direction joining two bulk octahedral sites. $V(R)$ (solid line) is the sum of the contributions a) (dots), b) (dashed line) and c) (dot-dashed line) described in the text. B: Fusion rate upper bounds at different pair energy E . The lozenges represent fusion from the bound states of $V(R)$

At equilibrium, the fusion rates of hydrogen isotopes in Pd are given by the points at lower energy in Fig. 4B (fusion from the ground state). These values are orders of magnitude smaller than the results of FP, J and S, and represent very accurate upper bounds to the true fusion rates in an equilibrium situation. On the other side, the results quoted by some authors who varied arbitrarily electronic mass and charge^{15,16} or the plasmon cut-off frequency¹⁷ are *parametrical* studies with scarce physical credibility. In order to explain realistically a fusion rate per pair of the order of $10^{-23} s^{-1}$ two approaches can be followed: i) to advocate a mechanism which involves energy concentration up to 160 eV or ii) to create a

situation in which the potential of Fig. 4A is strongly modified so to enhance the corresponding fusion rates. Using thermal equilibrium considerations an energy concentration as described in i) cannot be achieved in an assembly of fewer than about 1000 nuclei¹¹. This implies a long range cooperative effect that seems extraordinarily implausible. Some proposals have been advanced, however, in this sense¹⁸. An attempt to explore the possibility ii) has been reported in Ref. 13. Since the pair potential $V(R)$ strongly depends on the shape of the electronic charge density of the host metal (large spatial variations of the density on a short length scale may bring the reactant hydrogen nuclei to close distance) the fusion process at an ideal surface has been considered. That to model the largest spatial variation of the electronic density which is expected to occur in lattice configurations such as dislocations, cavities, defects, etc.. The pair potential $V(R)$ calculated with the nucleus 1 fixed in a surface octahedral site and the nucleus 2 at distance R in the vacuum is shown in Fig. 5A. A larger binding energy and a closer equilibrium distance is achieved with respect to the bulk case. The corresponding fusion rates, shown in Fig. 5B, are enhanced by up to 23 order of magnitude in the case of fusion from a bound state whereas no significant variation can be expected for the *in flight* processes. It is very difficult to imagine other mechanisms which could enhance the fusion up to the values obtained by FP, J and S.

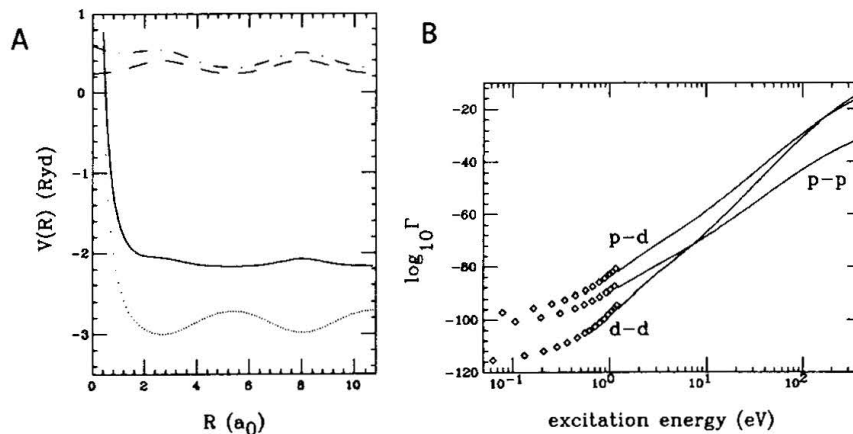
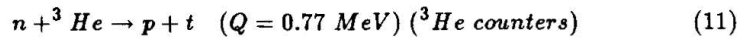
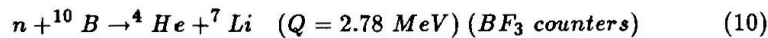


Figure 5. A: Shape of the pair potential $V(R)$ calculated along the direction joining two octahedral sites and crossing an ideal surface. $V(R)$ (solid line) is the sum of the contributions a) (dots), b) (dashed line) and c) (dot-dashed line) described in the text. B: Fusion rate upper bounds at different pair energy E . The lozenges represent fusion from the bound states of $V(R)$

3.2 n detection

Neutrons does not ionize but they can start nuclear reactions which produce charged fragments detectable by ionization detectors. Examples of nuclear reaction commonly used are:



These nuclear reactions have a large cross section only for slow neutrons ($1/v$ dependence of the cross section, where v is the n velocity see Fig. 6). It follows that detectors based on these reactions are highly efficient only for thermalized n ($E_n = 0.025 \text{ eV}$), and they must be surrounded by proton-rich material (moderator) to reduce, through multiple $n - p$ scattering, the energy of the incoming n . If the moderator is a scintillating material, then the n energy can be measured like in the case of the spectrometer used by J. Fig. 7 shows the pulse spectrum of an ${}^3\text{He}$ counter bombarded by a monochromatic n beam ($E_n = 1 \text{ MeV}$). We see the large slow neutron peak, corresponding to the energy of the proton from reaction of Eqn. 11 and a much smaller peak at higher energy in the case of non thermalized n . At lower pulse height there is a fast rising background induced by γ -rays and characteristic of this kind of gaseous devices. Conversely the $\gamma - n$ discrimination capability of a scintillation detector is based on the analysis of the shape of the scintillation pulse.

It should be known that n detectors like BF_3 and ${}^3\text{He}$ counters are rather unstable devices, mainly because of the impossibility of implementing coincidence techniques. An instructive example of the problems one can encounter using this kind of devices is represented by a set of experiments performed at Perugia from april 1989 to january 1990¹⁹ whose results were initially taken as evidence for burst of neutrons produced in CF. The data cover about 2500 hours of measurements using a moderated ${}^3\text{He}$ detector located near a S like cell. Temperature and pressure of the samples were precisely monitored and then their D_2 content was easily determined at each phase of the experiment. We used two different ${}^3\text{He}$ detectors, unfortunately never simultaneously, and both gave similar results. Initially only the n counting rate was monitored continuously and stored on a computer every second; later, also the analog pulses were analyzed and stored. Bursts of counts were detected with about one third of the samples used; these burst were lasting for few tens of seconds and sometimes they were very large. Fig. 8 shows one of the largest burst observed: in Fig. 8A the integral count is plotted as a function

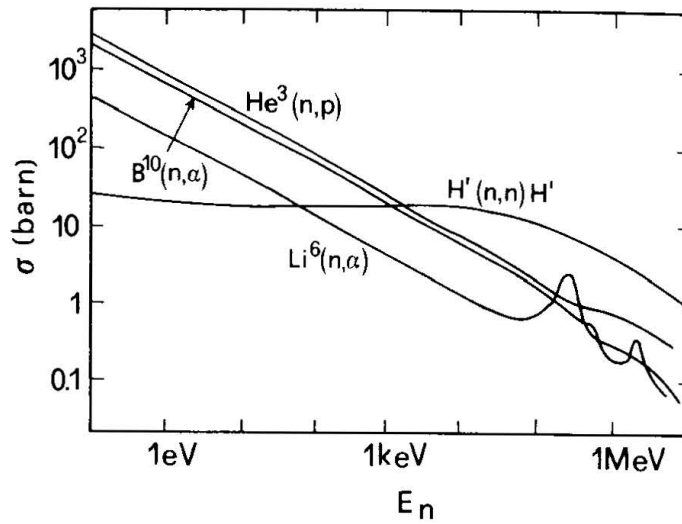


Figure 6. Cross sections for n -induced nuclear reaction as a function of neutron energy E_n .

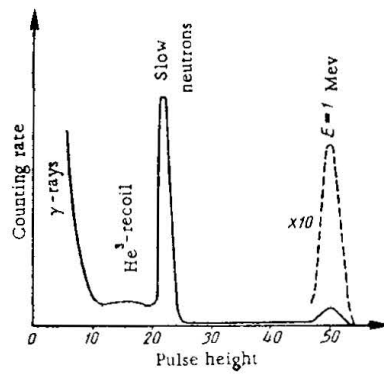


Figure 7. Pulse spectrum for ${}^3\text{He}$ proportional counters at $E_n = 1 \text{ MeV}$.

of time over a period of about 12 hours. The typical background level, monitored for days before and after the burst, is given by the slope of the curve during the first two hours (about 1 n/minute). The flux integrated over 1 second is shown in Fig. 8B. The time structure of the bursts were recorded by mean of a storage oscilloscope: an example is given in Fig. 9. These bursts were apparently related

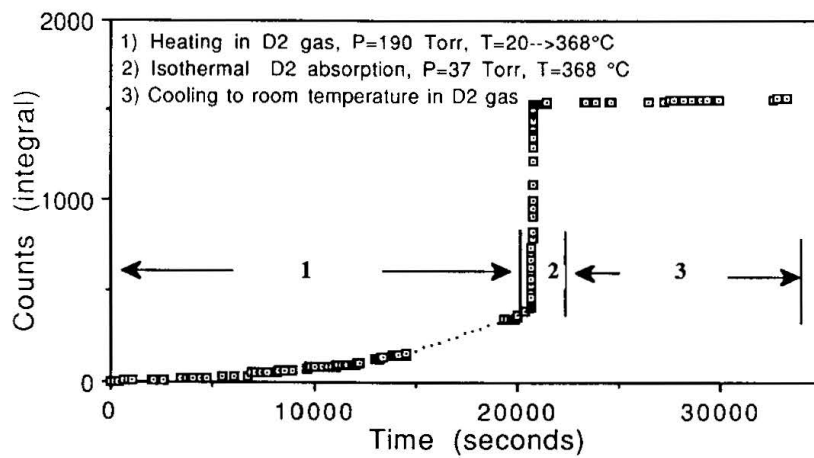
to absorption or desorption of D_2 by *fresh* (i.e. never exposed to D_2) *Ti* shaves. However, we soon realized that these counts were not due to n . A pulse analysis showed that, while the background was following the spectrum expected from an ${}^3\text{He}$ detector, the spectrum of burst pulses was different (Fig. 10). In addition, single pulses coming from the charge amplifier during bursts showed a shape very different from what was expected for n . These observations were indicating that we were observing counts not related to n emission. We then tried to understand which was causing the bursts. The possibility they were induced by sudden RF emission from the samples undergoing lattice deformations has been ruled out by a series of dedicated experiments, using a set of RF antennas. We were then forced to conclude that the bursts were unrelated to the physics of deuterated *Ti*. More likely they were due to preamplifier instabilities triggered randomly by humidity, temperature and possibly mechanical vibrations. A good rule coming from the experience is then to never trust counts from a single counter of this kind, in particular if one cannot look to the shape of the analog pulses. However this rule has been repeatedly violated in the first generation of CF experiments.

It is possible, but rather difficult, to measure the same n in two different detectors (coincidence), by detecting multiple elastic scattering with nuclei. By the measurement of the time difference between pulses, it is also possible to determine the n velocity, discriminating against γ induced background²⁰. The capability of making coincidence between different detectors, both in the *strict* sense of measuring twice the same n and in the *wide* sense of having two or more detector looking at the same time to the same source, is a very powerful way to improve the experimental sensitivity by reducing various kind of backgrounds. This capability was completely missing in the case of the FP and S experiments, and only marginally present in the case of J. In fact he performed coincidences between two signals coming from the same detector, but was trusting its performance over a rather long period of time while monitoring a small excess of n in the presence of a large experimental background.

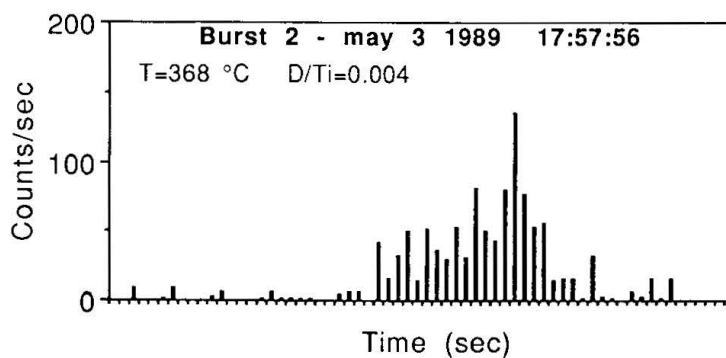
3.3 γ detection

When detecting γ in the (few) *MeV* range, the problem has two aspects, detection efficiency and energy resolution. Rather large *NaI* crystals may reach an efficiency of few per cent in this range with an energy resolution of about 50 *KeV*. More accurate (but smaller in size, i.e. less efficient) *Ge* detectors have a typical energy resolution of 0.5 *KeV*. A good energy resolution is important to distinguish a signal from the large background due to natural isotopes, characterized by well known spectral lines.

A very instructive example is given by the γ difference spectrum presented by FP for energies around 2.2 *MeV*. Near to the 2.224 *MeV* line corresponding to the deuterium recombination reaction there are two other lines (2.117 and



A



B

Figure 8. A: Integral rate observed at Perugia during D_2 absorption by Ti shaves. B: Rate integrated over one second.

2.204 MeV) corresponding to the decay chain of ^{214}Bi . Using a NaI detector, the second Bi line cannot be distinguished from the deuterium line. Then monitoring the γ background as did FP is meaningless, because the Bi contamination may change substantially from one place to another. When using a Ge counter the lines are easily separated as it is shown in Fig. 11 obtained at Perugia, and the difference in size of the deuterium peak when the cell is or is not operated represent a clean way to detect the presence of anomalous n production.

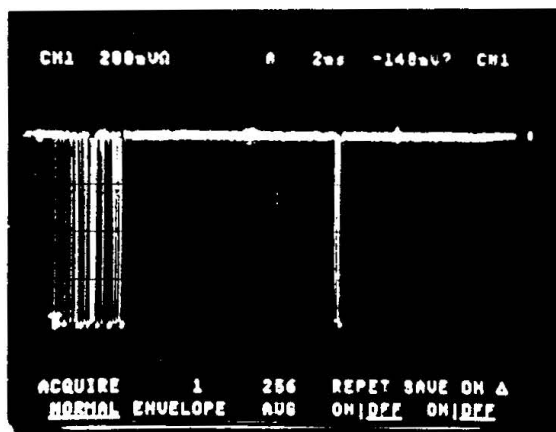


Figure 9. Time structure of one of the burst observed at Perugia; horizontal scale: 2 ms/square.

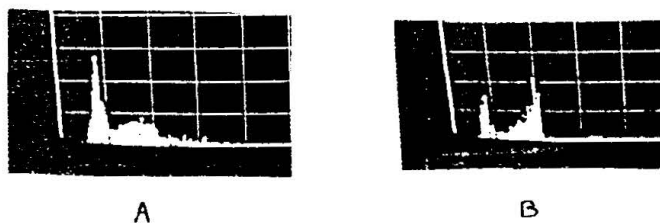


Figure 10. A: ^3He pulse spectrum collected during a burst. B: ^3He pulse spectrum during a background run.

3.4 ^3He , ^4He and t detection

The two He isotopes are stable and their presence can be detected only by mass spectrometry of cathodic samples before and after the operation of the cell. Since He is a rare gas, the detection of anomalously large quantities of it in the cathodes, would be an unambiguous indication for nuclear reactions. However this analysis is difficult, cannot be performed continuously during the experiments, and one should be aware of the possibility of accumulation of light elements in the cathodes during electrolysis.

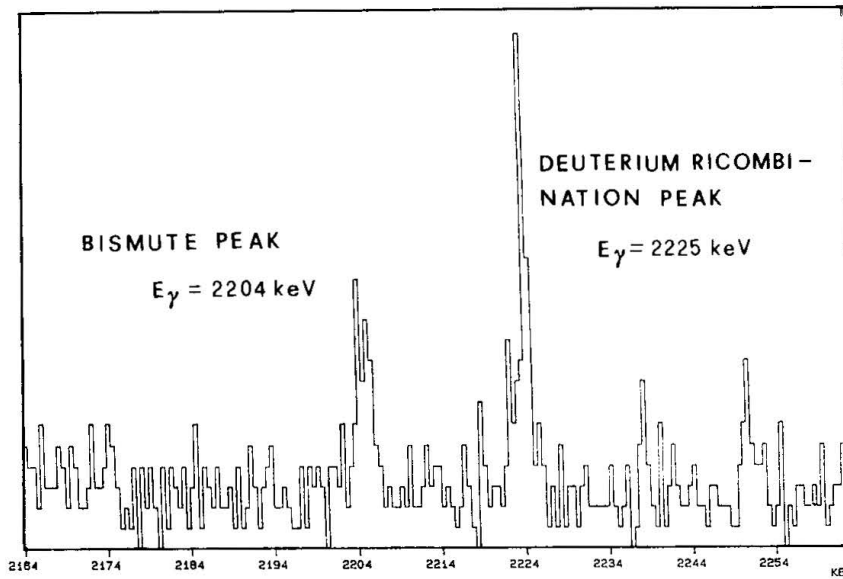


Figure 11. Background γ spectrum recorded at Perugia using a Ge detector.

Tritium is unstable can be detected by measuring the light produced when a sample of electrolyte is mixed with a liquid scintillator. One important remark is that heavy water is often contaminated with tritium, and different samples may have tritium contents that differs by orders of magnitude. Recalling that a cell consumes the electrolyte and that heavy water has to be added frequently, it is essential to monitor the tritium content both of the cell and of the samples used to refill it. In addition, electrolysis may change the equilibrium distribution of tritium in the electrodes and in the electrolyte. In conclusion, meaningful monitoring of the tritium content is not a trivial task.

3.5 Calorimetry

FP performed their calorimetric measurements in an *open* configuration, that is with exchange of gases and heat between the cell, the surrounding bath and the laboratory. Given the temperature of the system and its dissipation constant, the excess of power generated by the cell can be deduced by the input power, W_i , by the relation

$$W_i = I_E V_E (1 - f_r) \frac{\Delta H}{nF} \quad (12)$$

where I_E and V_E are the current and the voltage of the cell, f_r is the fraction of O_2 and D_2 that recombine at the cathode and $\frac{\Delta H}{nF}$ is the thermoneutral potential for water dissociation. In an open system, the temperature is not uniform over the cell if the stirring of the solution is not canceling the existing temperature gradients. FP were implicitly assuming a uniform temperature, justifying their assumption with the stirring induced by the turbulence of the gases produced. However, in this case one can expect effects depending on the current intensity. A better technique is to use an open cell, surrounded by a bath kept at constant temperature by an electrical heater whose power is known. The most accurate measurements make use of closed cells, where all the produced gas is recombined and measured, kept at constant temperature.

The experience accumulated by many groups in the last few months clearly indicates that making calorimetric measurements with an accuracy below about 10% is difficult; in any case the technique used by FP seems the most inadequate and exposed to errors. Their original results are then not accepted as reliable by the large majority of the scientific community. Surprisingly enough, in spite of these criticisms, FP went on making calorimetric measurements in the original way.

3.6 Curiosities

Pd cathode which surface has been activated by properly reversing the cell polarization or by *blackening*, is capable of adsorbing and desorbing deuterium or hydrogen very rapidly. If an activated cathode loaded with d is exposed to air, the atomic d desorbed from the cathode reacts immediately with oxygen making water; the cathode is violently heated by the energy released by the reaction (147.3 kJ/mole of d). In test experiments¹⁹ we were able to literally burn paper samples using this phenomenon. The possibility of this kind of *catastrophic* phenomena should be taken into account when discussing calorimetric measurement, in particular when operating the cells at high current. In this case in fact, the electrolyte turbulence is so strong that significant fraction of the cathode may be in contact with air most of the time and/or large quantities of oxygen be included in the electrolyte. In this condition the rate of d recombination may be significantly increased in a way that is difficult to quantify with precision. If it is not properly taken into account this effect could simulate anomalous heat production in the cell.

Due to their mass difference, D_2 and H_2 have different physical and thermodynamical properties. For instance D_2O and H_2O viscosity differs by about 30%, being $1.26 \cdot 10^{-2}$ and $1.009 \cdot 10^{-2}$ Poise respectively and also their thermoneutral potential (water dissociation) is different, being 1.527 and 1.481 V respectively. d and p diffuse at a different speed in metals (this fact is exploited to separate the two isotopes). Also their thermodynamical curves both in normal condition but

particularly in metals, are different. These differences in physical properties play an important role when making *blank experiments*. In fact it is not necessarily true that, when replacing D_2O with H_2O , the difference in behaviour in the two samples are due to nuclear phenomena. In addition we have seen that at low energies the fusion rates of Eqn. 4- 6 are comparable. Also 3H_2 and D_2 have different properties: in particular 3H_2 vapour pressure is lower than D_2 and that would clearly induce an increase of 3H_2 content in electrolytes exposed to air.

4. THE HOT DEBATE ON CF

The three discoveries discussed above started a CF *rush* all around the world that lasted through the summer 1989. The publicity given by the media to CF research was enormous, and the psychological pressure on the scientific community very important. Many groups started suddenly reporting positive findings but all were suffering the problem of reproducibility. Somewhat later, systematic studies were published, showing negative finding. In the following we briefly mention some of these experiments, commenting on their peculiarities.

One of the first confirmations was obtained in the repetition of the J experiment at the underground italian laboratory of Gran Sasso²¹ by a group including S. Jones himself. They used two liquid scintillators $NE - 213$, one located near the cell and the other 8 m away; n were separated from the large γ background induced by the natural rock radioactivity by shape analysis. Background runs were taken before and after the cell operation and the background was subtracted from the rate measured during cell operation. In the data reported in the paper, the detector near to the cell showed an increase in rate (two to three standard deviations after background subtraction) during three subsequent runs, each lasting about one hour. After about three hours the rate came back to the background value. The n energy distribution was well described by a Monte Carlo calculation for 2.45 MeV n taking into account the details of the experiment. The rate reported was about 20% lower than the original J result, and, due to the high γ background, also the statistical significance of this new experiment was not overwhelming.

Another italian group lead by P. Perfetti²² performed a variant of the FP experiment. They looked for n emission from electrochemically D_2 loaded Pd and Ti wires, during heating cycles performed outside the cell. A current of 10 A passed through the wire reaching temperatures higher than $100^\circ C$. The n detector consisted in an array of 30 3He detectors all connected in parallel. This configuration seems rather unpractical because random drifts of a single counter would affect the performance of the full apparatus. They recorded the total counting rate integrated every minute. Following this procedure the authors claimed to have been able to repeatedly measure rates above the background ($2 n/minute$)

for short periods starting about 2 minutes after the current treatment. However the published data (Fig. 12), which represent only a fraction of the run performed, show a rather weak effect, reaching at most 3 standard deviations. The authors also reported the result of measurements performed during the operation of the cell; they do not see significant variations of the n counting rate but their sensitivity was about a factor 30 below the original J result.

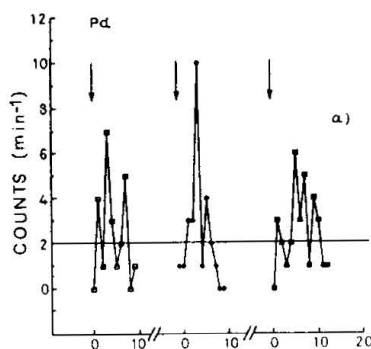


Figure 12. Neutron counts integrated every minute for three different experiments performed on Pd charged with D_2 and subjected to one minute current bursts. The arrows refers to the beginning of heating (Ref. 22).

Unique events were reported by a group working at the Gran Sasso laboratory²³. During the operation of a cell they observed a 0.6 s long shot event during which the counting rate increased hundredfold simultaneously in a NaI γ detector, in a bare 3He detector and in another 3He detector surrounded by paraffin. However, it seems very hard to believe that these burst are due to detection of real particles like n or γ . In fact NaI crystals do not detect n and, in addition, if these particles were n , the moderated 3He counter should have counted much more than the other and not a factor two less. In addition, for proper threshold settings, 3He detectors are rather insensitive to γ up to few hundreds of KeV. Even in the case of an intense burst of high energy γ ($E_\gamma \geq 0.5 - 0.7 MeV$), the n counters are expected to be much less efficient than a NaI crystal, while the rates reported were similar within a factor of two. Since this observation has never been confirmed since then, other less appealing effects like large fluctuations in the power line inducing spurious counting on the electronics, cannot be ruled out in this case.

More intriguing is the observation of another unique event by a group of

the Department of Chemistry at the University of Rome²⁴. They operated a FP like cell and monitored the current and the temperature of the cathode, made of sintered *Pd* powder. In addition an ^3He REM counter, was placed near to the cell. They reported a single event (Fig. 13) where the cathodic temperature suddenly rose to above 100°C , after which the safety system switched off the cell. *Simultaneously* the *n* counter did record a spike of 36 *n* in about 4 minutes compared to a background of 3 *counts/hour*. The data were written on a chart recorder. This event is very interesting in view of the large heat released in *coincidence* with the detection of a *n* burst. However the authors found that assuming conventional nuclear theory the *n* flux detected is about 9 orders of magnitude below what would be necessary in order to explain for the heat released. The authors also claim having detected a large increase of the tritium content of the solution after the event with respect to other blank experiments they performed. If they assume the tritium was produced during the heating, they are still three orders of magnitude too low to explain the heat observed. However, assuming that the tritium density in the solution is a factor 100 smaller than its density inside the cathode, as suggested in Ref. 25, they are able to get close to balancing the heat observed and the energy released by nuclear products. This analysis implies an asymmetry of about 8 orders of magnitude between the two reactions of Eqn. 4, in total disagreement with the standard nuclear theory at low energy which predicts about the same branching ratio in the two channels. In spite of the detailed knowledge of the cathodic structure and control of the experimental procedure, the group was unable to reproduce this extraordinary event during the year following the first experiment.

Is it possible to find an explanation for the observed effects that does not take into account nuclear reactions? At first view it could seem impossible, given the simultaneous measurement of heat and neutrons. In our opinion however, the possibility of sudden heating of the cathode due to anomalous O_2 and D_2 recombination in the vicinity of the cathode cannot be completely ruled out. Even if, as it is stated in the paper, at the time when the heating started the cathode was covered by (only) 6 *mm* of solution (at the time when the heating took place nobody was present, and later the top of the cathode was found above the solution level, with some of the water in the exhaust line), it is well known that in FP cells the turbulence due to gas production at the electrodes is very strong. It is then possible that some fraction of the cathode surface had been exposed to the mixture of D_2 and O_2 present on the top of the cell, starting a recombination reaction that might have developed in an explosive way. This violent chemical reaction, followed by the switching off of the cell, might have induced spurious counts on the adjacent neutron counter (the kind of counter used is known to be sensitive to electrical and acoustical noises) in *coincidence* with the heating of the cathode. We like to stress that the above scenario is just a speculation suggested to us by direct experience with *n* counters and FP like cells. However we feel it is important, in the presence of extraordinary, irreproducible events, to be able for-

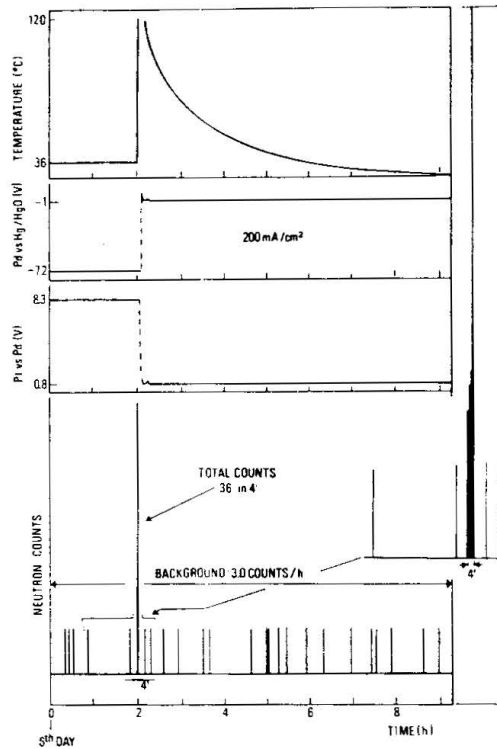


Figure 13. Paper recorder traces of the experimental variables during the event. An enlarged view of the recorder neutron counts in a time interval around the event is also shown (Ref. 24).

ulate alternative hypothesis, to the "there-is some-extraordinary-new-physical-effect-that-take-place-only-when- some-unknown-parameters-take-very-particular-values" hypothesis. In particular we are not able to explain, in our scenario, the large increase of tritium measured by the authors.

Experiments performed by the Texas A. M. group²⁶ focussed on heat production in FP like cells and on tritium measurements. The results obtained clearly suggest that lithium plays an important role in producing excess of heat, and experiments performed using H_2O instead of D_2O did not show anomalous heat production (*sic*). Also the amount of tritium in the solution was monitored and its density was observed to increase with time; it is not clear from the paper if this effect might be due to the difference between the vapour pressure of D_2 and 3H . However, while the excess of heat was detected after few hours of cell operation at high current (after many hours at low current), the tritium increase was detected only after 2 – 6 weeks of operation. After an analysis of the possible chemical

sources for anomalous heat production, the authors conclude it is due to nuclear reactions. Unfortunately n and γ were not detected during these experiments and then this conclusion is not completely satisfactory. This experiment, although incomplete, using open and closed calorimetry at constant temperature is possibly the most important result regarding heat production in FP like cells.

The cover of Nature of July 6th 1989, was titled *No evidence for cold fusion neutrons*. The title was referring to two papers, one of M. Gai and collaborators²⁰ and the other of N. S. Lewis and collaborators²⁷ in which limits on heat, n and γ production in FP like cells were given. Both groups had reported their findings at the APS meeting that took place at the beginning of May. The first paper gave a limit for n flux about 50 times lower than the result quoted by J and about 6 orders of magnitude lower than the FP value. This group used a sophisticated n coincidence technique, detecting the same n on two different counters and measuring its time of flight. The γ production was monitored using *NaI* detectors; no excess was found in particular at energies corresponding to the known nuclear reactions. At the end of May, S. Jones agreed to repeat his experiment together with the Gai group and later in the summer they reported their negative findings to the Department of Energy panel.

The second paper contains, in addition to limits on n and γ production (around 2.2 MeV they do not find the FP γ peak using a very precise *Ge* detector), a detailed discussion of the problems connected with calorimetry in electrochemical cells. For instance, there is a dependence of the $O_2 + D_2$ recombination rate from the current in the cell, the temperature measured in the cell depends on the amount of stirring, the coefficient of heating of the cell varies with time and depends on the cells current. All these effects were neglected in the original FP paper. In conclusion this study shows that it is extremely difficult to perform calorimetry on cells with an accuracy better than about 6%, an accuracy that is comparable with the effects claimed by FP.

The Harwell group performed a long and costly series of experiments, initially with the direct help of M. Fleischmann. They announced their negative results in a press conference (on 15th June) and later published a paper on Nature²⁸. This study is probably the most accurate and detailed ever performed. They repeated the FP calorimetry, finding many uncertainties not taken into account in the original work, like a dependence of the calibration constants on the electrolyte level and the need for careful stirring in order to trust the temperature measurement. In addition, performing accurate closed cell calorimetry they did not measure any significant excess of heat. n was measured by *BF₃* counters placed in 5 concentric shells and read independently; drift in the counting rate of one or two counters was observed but never a coincidence between a significant number of them was recorded. The γ spectrum was measured with a large *Ge* detector but no excess was found at energies corresponding to nuclear reactions

like deuterium radiative recombination. The tritium content was monitored continuously showing a marked (50%) increase with time due to catalytic enrichment of the solution. The group performed an impressive number of experiments using different geometries and cathodic materials, varying all the possible parameters within reasonable values and in all condition was unable to reproduce the results claimed by FP.

The results published by Nature had a strong impact on the scientific community suggesting to many scientists the possibility that part (if not all) of the CF saga was due to wrong results amplified by media. These measurement were eagerly awaited and the negative findings resulting from the systematic and careful work of large groups of experts represent a turning point in CF history.

Experiments were also done to verify the claim of the Frascati group. In an interesting paper, Segre and collaborators¹⁸ present a systematic scan of the phase space of deuterated Ti , looking for anomalous n production rate in the presence of phase transitions. In Fig. 14 a typical scan is shown. Using both 3He BF_3 n counters they found a negative result, setting limits on n production much lower than S and FP but somewhat larger than J. This group also reported the erratic behaviour of a BF_3 counter triggered by a power spike and lasting for many days, as an example of the danger involved when performing experiments relying on only one detector.

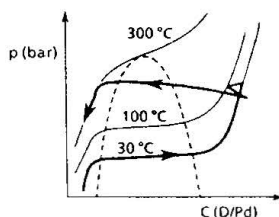
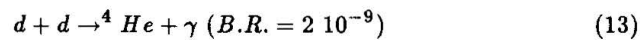


Figure 14. Example of the thermodynamical cycles performed in the experiment discussed in Ref. 18.

The Culham-JET-Harwell group also performed S type experiments, monitoring the n flux with 4 counter banks, two monitoring the background and two the S like cell. They also found negative results²⁹ both during cooling and heating experiments, but observed occasional drift in time of some of the counters lasting in some cases for hours, but never in coincidence. They were unable to explain these misbehaviours, but suspected they was related to partial electrical breakdown in the insulating material of the HV connections.

Accurate experiments were also performed by a collaboration involving SIN, Garching, ETH and Wien ³⁰ involving both accurate calorimetry, search for ³He and ⁴He and *n* through mass spectrometry and γ detection. They also performed routinely blank experiments using H_2O . The technique used for *n* detection was the same used at Gran Sasso in the repetition of the J experiment, but they place an upper limit slightly lower than the flux reported at Gran Sasso by Bertin and collaborators. For what calorimetry is concerned this group performed closed calorimetry at constant temperature, placing an upper limit of 0.1 W on the excess of heat observed. In addition the analysis performed on the cathodes allows to place a limit on the presence of ⁴He that is about 6 orders of magnitude below what would be needed in order to explain the heat detected by FP in terms of the *n* free reaction



Negative results have also been reported by the Milano group lead by E. Fiorini using both FP (electrolysis) and S (gas) cells ³¹. In the first case they tried (and failed) to induce *n* production by irradiating the cell with α or γ sources, testing the possibility that irreproducible *n* bursts are generated by cosmic rays showers. In the second case using 4 BF₃ counters they were able to put an upper limit on *n* production per *d* pair of $2 \cdot 10^{-24} \text{ s}^{-1}$.

Some experiments were reporting the observation of totally irreproducible *n* bursts using counter(s) located near FP or S like cells ³². In particular the S group reported the observation of a burst seen *almost* in coincidence by two BF₃ counters during an heating phase ³³. However in view of all the problems encountered with this kind of *n* detectors these claims really need further confirmation and systematic studies.

5. SCEPTICS AND BELIEVERS

If we try to summarize the situation at the beginning of 1990, we realize immediately that negative results on CF are overwhelming with respect to positive results. Most of the positive results were obtained with poor experimental facilities without too many precautions and redundancy in the detecting system. However, it is impossible to demonstrate that all the reported positive results are due to experimental errors and some doubts remains that CF is really taking place in metals at a detectable level.

In any case, some general consideration are possible: *a*) the original experiments that claimed positive results were simple, not systematic, not redundant;

more powerful experiment, that are redundant and systematic do not see any effect; *b*) the original data were affected by various problems and their significance is very debatable. Evidence of some strange effects has been since then obtained in new experiments often different from the originals, but never in a convincing way; *c*) accurate and simultaneous measurements of heat and nuclear products (n , γ , t , ${}^3\text{He}$, ${}^4\text{He}$) are difficult and many kind of errors are possible.

In conclusion, one year after the FP press conference, perspectives for CF seem rather bad. The very high economical and social interests related to this subject transformed the scientific issue in a problem of faith. With time the scientific community has been divided in skeptic and believers and there are no indications that the situation will ever change. Very few groups are still doing serious research on the subject and the reports of high quality results are now infrequent. Conferences on the subject are organized mainly by believers with the risk of not giving adequate relevance to groups reporting negative results. The way CF developed suggest strongly it is a case of pathological science³⁴. If it is really the case will become more clear in the next few years. In any case the CF saga will be remembered for a very long time.

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REFERENCES

1. D. R. O. Morrison has called our attention on this Hume's statement.
2. M. Fleischmann and S. Pons, *J. Electroanal. Chem.*, **261**, 301 (1989).
3. R. L. Garwin, *Nature* **338**, 616 (1989).
4. A. De Ninno et al., *Europhys. Lett.*, **9**, 221 (1989).
5. J. D. Jackson, *Phys. Rev.* **106**, 330 (1957).
6. Ya. B. Zel'dovich and S. S. Gershtein, *Usp. Fiz. Nauk* **71**, 581 (1960) [*Sov. Phys. Usp.* **3**, 593 (1961)].
7. C. De W Van Siclen and S. E. Jones, *J. Phys. G* **12**, 213 (1986).
8. W. A. Fowler, G. R. Caughland and B. A. Zimmerman, *Annual Rev. of Astr. and Astrophys.* **5**, 525 (1967).
9. S. E. Koonin and M. Nauenberg, *Nature* **339**, 690 (1989)
10. N. R. Arista, A. Gras-Martí and R. A. Baragiola, *Phys. Rev. A* **40**, 6873

- (1989)
11. A. J. Legget and G. Baym, *Phys. Rev. Lett.* **63**, 191 (1989).
 12. E. Wicke and H. Brodowsky, *Hydrogen in Palladium and Palladium Alloys*, in *Hydrogen in Metals II*, G. Alefeld and J. Völkl, eds., (Springer-Verlag, Berlin 1978).
 13. F. Marchesoni, C. Presilla and F. Sacchetti, *Europhys. Lett.* **10**, 493 (1989).
 14. K. Sohlberg and K. Szalewicz, *Phys. Lett. A* **144**, 365 (1990).
 15. J. Rafelski, M. Gajda, D. Harley and S. E. Jones, University of Arizona preprint AZPH-TH/89-19.
 16. F. Parmigiani and P. G. Sona, *Il Nuovo Cimento D* **11**, 913 (1989).
 17. M. Baldo and R. Pucci, in *Understanding Cold Fusion Phenomena*, Conference Proceedings vol. 24, R. A. Ricci, E. Sindoni and F. De Marco, eds., SIF Bologna (1989).
 18. S. E. Segre, S. Atzeni, S. Briguglio and F. Romanelli, in *Understanding Cold Fusion Phenomena*, Conference Proceedings vol. 24, R. A. Ricci, E. Sindoni and F. De Marco, eds., SIF Bologna (1989); E. Tabet and A. Tenenbaum, *ibidem*.
 19. R. Battiston, R. Borio, F. Mazzolai, G. Ambrosi, C. Costa and P. Scampoli, unpublished.
 20. M. Gai et al., *Nature* **340**, 30 (1989).
 21. A. Bertin et al., submitted to *Nuovo Cimento*.
 22. P. Perfetti et al., in *Understanding Cold Fusion Phenomena*, Conference Proceedings vol. 24, R. A. Ricci, E. Sindoni and F. De Marco, eds., SIF Bologna (1989).
 23. F. Celani et al., in *Understanding Cold Fusion Phenomena*, Conference Proceedings vol. 24, R. A. Ricci, E. Sindoni and F. De Marco, eds., SIF Bologna (1989).
 24. D. Gozzi et al., to be published in *Il Nuovo Cimento A*.
 25. N. J. C. Packham et al., *J. Electroanal. Chem.* **270**, 451 (1989).
 26. A. J. Appleby et al., in *Understanding Cold Fusion Phenomena*, Conference Proceedings vol. 24, R. A. Ricci, E. Sindoni and F. De Marco, eds., SIF Bologna (1989).
 27. N. S. Lewis et al., *Nature* **340**, 525 (1989).
 28. D. E. Williams et al., *Nature* **342**, 376 (1989).
 29. G. M. McCracken et al., Culham preprint CLM-P864, (1989).
 30. J. P. Blaser et al., *Chimia* **43**, 262 (1989).
 31. A. Alessandrello et al., in *Understanding Cold Fusion Phenomena*, Conference Proceedings vol. 24, R. A. Ricci, E. Sindoni and F. De Marco, eds., SIF Bologna (1989).
 32. M. Di Fino et al., in *Understanding Cold Fusion Phenomena*, Conference Proceedings vol. 24, R. A. Ricci, E. Sindoni and F. De Marco, eds., SIF Bologna (1989); B. Emmoth et al., *ibidem*.
 33. A. De Ninno et al., in *Understanding Cold Fusion Phenomena*, Conference Proceedings vol. 24, R. A. Ricci, E. Sindoni and F. De Marco, eds., SIF

Bologna (1989).

34. D. R. O. Morrison, CERN/EP 90-36, 15 March 1990.