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Background to Cold Fusion: the Genesis of a Concept

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The scheme of research which led to the start-up of the project now known as “Cold Fusion” is illustrated by Fig. 1. We note that it is commonly believed that there is absolutely no way of influencing Nuclear Processes by Chemical means: therefore, any results that demonstrate that this might be possible must be due to faulty experimentation, delusion, fraud etc. However, any enquiry as to the *experimental* foundation of the first statement in Fig. 1 is normally met by the response: “*because quantum mechanics, Q.M., shows that this is so*”¹ (see further below). We are driven to the conclusion that this first statement is just part of the belief-system of Natural Scientists and we naturally also have to ask the question; “what conclusion would we draw if we subject the statement to the dictates of Field Theory?”^{2,3}

In the 1960’s we started a series of research projects aimed at answering the Question; “can we find illustrations in Chemistry (especially Electrochemistry) of the need to invoke the Q.E.D. paradigm to explain the results obtained?” The opposition we encountered in applying this line of reasoning to the first of these projects,

(i) the kinetics of fast reactions in solution at short space-times (2×10^{-22} cm.s to 3×10^{-12} cm.s), see Fig.1, convinced us that such research had to be carried out using “hidden agendas”, see Fig. 2

¹ Which coincidentally explains why I did not start this project at any time during the period 1947-1983

² It is well known that the Quantum Mechanical Paradigm is incomplete and that the inconsistencies produced by this paradigm are removed by Quantum Field Theory, Q.F.T., (or Quantum Electrodynamics, Q.E.D., for applications to ordinary matter).

³ Critiques based on the application Q.M. to dilute plasmas are usually extended to cover fusion in condensed matter. The statement “no neutrons, no fusion” is a typical example of this reasoning. We note that an excellent text book covering the application of Q.E.D. to condensed matter is available (1).

Chemistry \longleftrightarrow Nuclear Physics

Because Q.M. says so
But what about Q.E.D.?

Q.E.D. in Chemistry;

- (i) Kinetics of fast reactions in solution at short space-times
- (ii) Voltage-gated transmembrane ion currents
- (iii) Kinetics of nucleation and phase growth
- (iv) surface x-ray diffraction
- (v) wall-phase turbulence

Coherent and Incoherent Structures

Suppose this is also true for Nuclear Physics

Compression and shear of metal lattices
Bridgman 1930's
"Cold explosions"

Chemistry
Physics

Compression of Metal Deuterides

Chemistry

Use of eV perturbations to create structures at the G.E.V. level

Fig. 1 The scheme of research which led to the start-up of the "Cold Fusion" project.

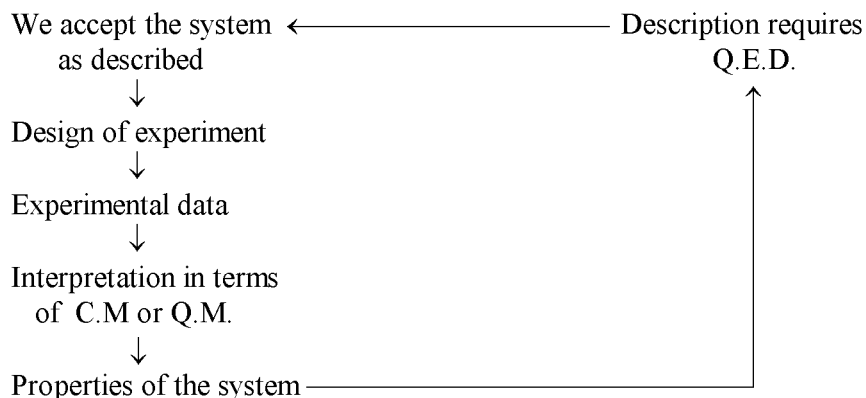


Fig. 2 The “hidden agendas” of the research projects.

Thus, using such agendas, the systems are investigated within the accepted models of C.M. or Q.M.: the influence of many-body effects and of Q.E.D. had to emerge from the interpretation of the results. It will be evident that this particular strategy had the advantage of avoiding the presupposition of the importance of Q.E.D.; for example, it might well have turned out that such effects were unimportant and/or not measurable. The strategy also had the advantage of avoiding the premature criticisms of the multitude of scientists opposed to the application of Q.E.D. to condensed phase systems ! The overall aim was the illustration of the developments of inconsistencies arising from the purely atomistic view of Quantum Physics: we need to answer the question whether the description of Nature requires collective dynamics (*field* dynamics in the physical jargon) for the analysis of the seemingly individual phenomena.

One conclusion which followed from these investigations of (i)-(iv) was that condensed phase systems developed structures having dimensions lying between $\sim 100-1000\text{\AA}$, structures that played a crucial rôle in the behaviour of the systems.⁴

At a later date, we adopted the terminology of Q.E.D. Coherence (1) by describing the separation of the bulk material into Coherent and Incoherent Domains. We will confine attention here to just one of these topics (i)-(v) namely, surface X-ray diffraction, (iv), as this methodology gives direct information about the structure of electrolyte solutions (topics (i) and (ii) were described briefly in the Proceedings of the Ninth International Conference on “Cold Fusion”,(2)). Two factors made such a seemingly impossible project feasible, (3), The first was the advent of position sensitive single photon counting, Figs. 3A and B, the second was the use of modulation techniques to enhance the selectivity towards surface diffraction, Figs. 4A and B. Fig. 3A illustrates the details of the methodology originally proposed, the use of a multi-wire proportional counter in combination with a bright rotating anode source. However, in view of restrictions in the funding, the project was eventually carried out with a linear position sensitive detector in combination with a fixed anode source. We estimated that this caused a degradation in the performance by a factor in the range 100 to 1000. Fig. 4A illustrates the diffractogram obtained when using a thin layer cell containing an electrode consisting of a thin film of silver. We observe the (111) and (200)

⁴ It follows, therefore, that the effects of these structures are most readily investigated by carrying out experiments on systems having such small dimensions.

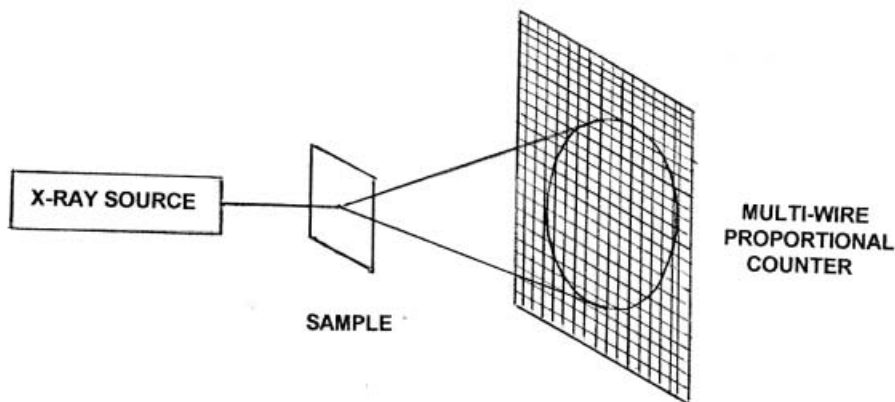


Fig. 3A. Proposed experiment for measuring x-ray diffraction.

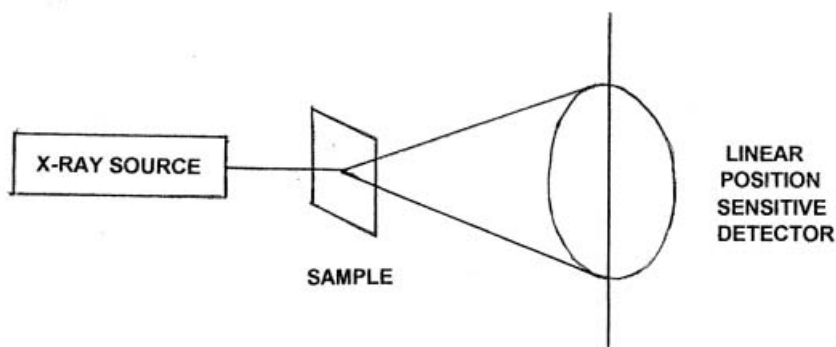


Fig. 3B. Execution of the experiment on surface x-ray diffraction.

diffractions of silver superimposed on a broad background due to diffraction from the aqueous solution in the cell. Clearly, such diffractograms do not give any useful information. However, modulation of the potential between the values for the observation of the silver surface alone and that for underpotential deposition of a single layer of lead (4) coupled to long data acquisition and the use of a 15-point triangular smoothing window, gave the difference diffractogram illustrated in Fig. 4B.⁵ The main features seen in Fig. 4B are a loss of order around $2\theta = 17^\circ$ and 44° , a differential shaped feature centred around $2\theta = 26^\circ$ and, associated with this a differentiation of the peak at $2\theta = 38^\circ$ due to the Ag (111) reflection. The loss of structure at 44° is particularly interesting as this corresponds to the Ag (200) peak which is reduced in magnitude as the lead monolayer is formed. From optical and voltametric studies it has been concluded (4) that deposition of lead on Ag (100) surfaces corresponds to an higher order phase transformation which would lead to a marked change in the structure factor but no clearly defined diffraction peak. By contrast deposition on Ag (111) surfaces has been assigned to a first order phase transformation. It is logical therefore to assign the differential peak to the formation of an incommensurate monolayer on the (111) facets of the polycrystalline silver film electrode.

⁵ We note that it is possible to observe such difference diffractograms notwithstanding the degradation of the performance implied by using the instrumentation illustrated in Fig. 3B rather than that originally envisaged, Fig. 3A.

However, the most surprising results of the investigation were the observation of the differential peak centred around $2\theta = 26^\circ$ and the loss of structure around $2\theta = 17^\circ$. The first is most logically attributed to a lengthening of the O-O distance in the water near the electrode surface as the lead monolayer is formed (the large peak centred around $2\theta = 26^\circ$ is generally attributed to hydrogen bonded water). We can judge this change in structure to involve the equivalent of 20-50 monolayers of water. The second, corresponding to a nearest neighbour distance of 6.8\AA is most likely due to a change in the ion distribution ; we can see that the distribution of ions is much more highly ordered than that predicted by present day models of electrolyte solutions.

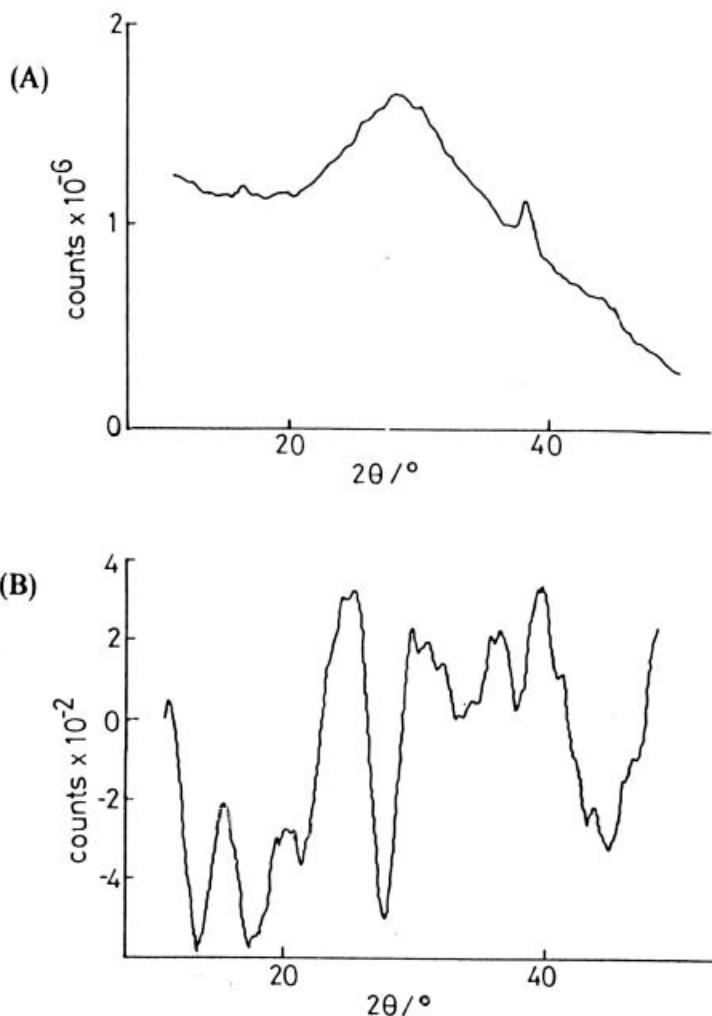


Fig. 4. In situ x-ray diffractograms for a thin silver electrode in $100\text{mM Pb}(\text{ClO}_4)_2 + 0.5\text{M NaClO}_4 + 1\text{mM HcClO}_4$.
 A) diffractogram obtained after 50h at -400mV
 B) difference diffractogram (diffractogram at -400mV minus that at -100mV) after 100h modulation at 10^2Hz .

The main conclusion which follows from the investigation is that we cannot achieve a complete description of the system in terms of the left-hand-side of the “hidden agendas” of Fig. 2. Such a description must clearly involve strong interactions between all the species in the interfacial regions i.e. we have to consider the many-body problem within the context of field theory. We note that the application of Q.E.D. to the solvent and electrolyte solutions shows that the solvent (water) must be divided into two domains, one in which the solvent is highly structured (the coherent regions) and the second (the incoherent regions) in which the solvent is disordered; electrolyte is expelled from the coherent into the incoherent regions where it in turn forms ordered domains (1), (2), (5), (6). This line of reasoning prompts us to attribute the changes in the diffractogram, Fig. 4B, at $2\theta < 26^\circ$ to the selection of coherent domains by the underpotential monolayer of lead on the silver substrate coupled to an enhancement of the scattering from the incoherent regions.

By 1983 we had reached a position where it seemed logical to ask the next question posed in Fig. 1: suppose the division of solid phases into coherent and incoherent structures was also true for Nuclear Physics. If that was so, then would we be able to use the perturbations at the eV level (characteristic of chemical changes) to create coherent structures at the GeV level? Furthermore, would we be able to observe nuclear processes in such structures? Of course, in posing these questions we were also influenced by other relevant work such as the interpretation of Mossbauer spectra, see (1) and, especially, the observations of “Cold Explosions” by Bridgman in the 1930’s (7).⁶ Furthermore, we knew that the absorption of hydrogen isotopes can lead to a similar fragmentation. This then was the background which led to the specification of the first system for our investigations; the highly forced electrolytic charging of palladium cathodes with deuterons.⁷ It is appropriate here to comment also on the chosen methodology, the measurement of the thermal balances using calorimetry (8), (9). It was clear at that time that the favoured methods of Nuclear Physics could only be applied to the systems we had in mind following many modifications and with numerous restrictions. Furthermore, there were ambiguities in the interpretation of some of the results which had been obtained.⁸ On the other hand, the measurement of thermal balances would be straightforward and inexpensive (an important factor as we were carrying out the initial investigation at our own expense) and such methods could achieve a high sensitivity and be reasonably free from ambiguities (however, see further below).

We therefore embarked on this project but without any expectation that we would obtain definitive results. The Pd/D system was investigated (coupled to the use of the Pt/D system as a suitable “blank”). However, the outcome was radically different to our expectations: the generation of excess enthalpy without any significant formation of the fusion products produced in dilute high temperature plasmas.⁹ When the project reached the public domain, (8), (9), it

⁶ Intense compression and shear of lattices can lead to their fragmentation into small particles in which the high energy of the initial system is converted into kinetic energy of the fragments. We believed (and still believe) that this is a process which can only be explained by Q.E.D.

⁷ Although we could see that the results obtained opened the way for very wide-ranging investigations, our own work never progressed beyond this initial concept. We described the resources required for the much wider investigations as equivalent to a Manhattan II project; this was true even for the much more limited scope of a project which remained restricted to the charging with deuterium of palladium based host lattices.

⁸ See, for example, the interpretation of the tracks due to $^3\text{T}^+$ and $^1\text{H}^+$ in a Wilson Cloud Chamber (10), (11), an experiment carried out following the discovery of the first hot fusion reaction (12).

⁹ The formation of ^3T could be shown to be far above that which could be attributed to isotopic separation with any reasonable choice of separation factors; the formation of neutrons appeared to be confined to the non-steady state of

became clear that our expectations with regard to the use of calorimetric methods could not be realised and it is important at this stage to ask why this might have been so ?

One of the causes is undoubtedly the inherent irreproducibility of the phenomenon. We believe that the process which we have observed takes place in the bulk of the electrode, (8), (9), as mediated by the surface reactions. We can see that this immediately builds in inherent irreproducibilities into the system. Moreover, it has not generally been appreciated that although it is relatively straightforward to produce palladium electrodes, the metallurgy of palladium is difficult leading to the use of unsatisfactory (and inadequately specified) electrode materials.

However, the major reason is undoubtedly the fact that the calorimetry of electrode reactions has not emerged as a viable methodology (it remains confined to a number of sub fields such as the measurement of self-discharge in batteries). Papers dealing with the precision and accuracy of calorimetric methods and comparisons of various studies¹⁰ are rejected outright by Journal Editors and referees^{11, 12} so that the study of calorimetry does not advance. The rather wild statements which have been made in the literature about the precision and accuracy of calorimetry as applied to the Pd-D system have undoubtedly also been due to the complexities of excess enthalpy generation in this system. Thus, it does not appear to have been realised that it is impossible to calibrate any calorimetric system if this is subject to fluctuating sources of enthalpy generation. Such calibrations require at the very least a constancy of the rate of excess enthalpy generation but, better, a zero value of this rate. It follows, therefore, that such calibrations must be carried out when using suitable “blank” systems. The intervention of “positive feedback” in the Pd-D system is especially troublesome in this regard. (14) We illustrate this point by two calibrations carried out by the New Hydrogen Energy Group in Japan, Figs. 5A and B. The onset of “positive feedback” leads to a delayed approach to the steady state following the application of the calibration pulse and a delayed decay to the base-line following the termination of this pulse, Fig. 5B. Not surprisingly, attempts to derive the true heat transfer coefficient from calibrations subject to such effects lead to the impossible result that the true heat transfer coefficient is smaller than the lower-bound value e.g. see (15). As we have already noted, exact evaluations of the precision and accuracy of the instrumentation require experiments using “blank systems” (e.g. see (13)).

operation of the cells; the formation of ⁴He could be detected but could not be related to the magnitude of the excess enthalpy generation.

¹⁰ Admittedly with regard to studies in “Cold Fusion”

¹¹ However, the usual outcome is that such papers are simply returned to the authors without comment.

¹² For one such study see (13).

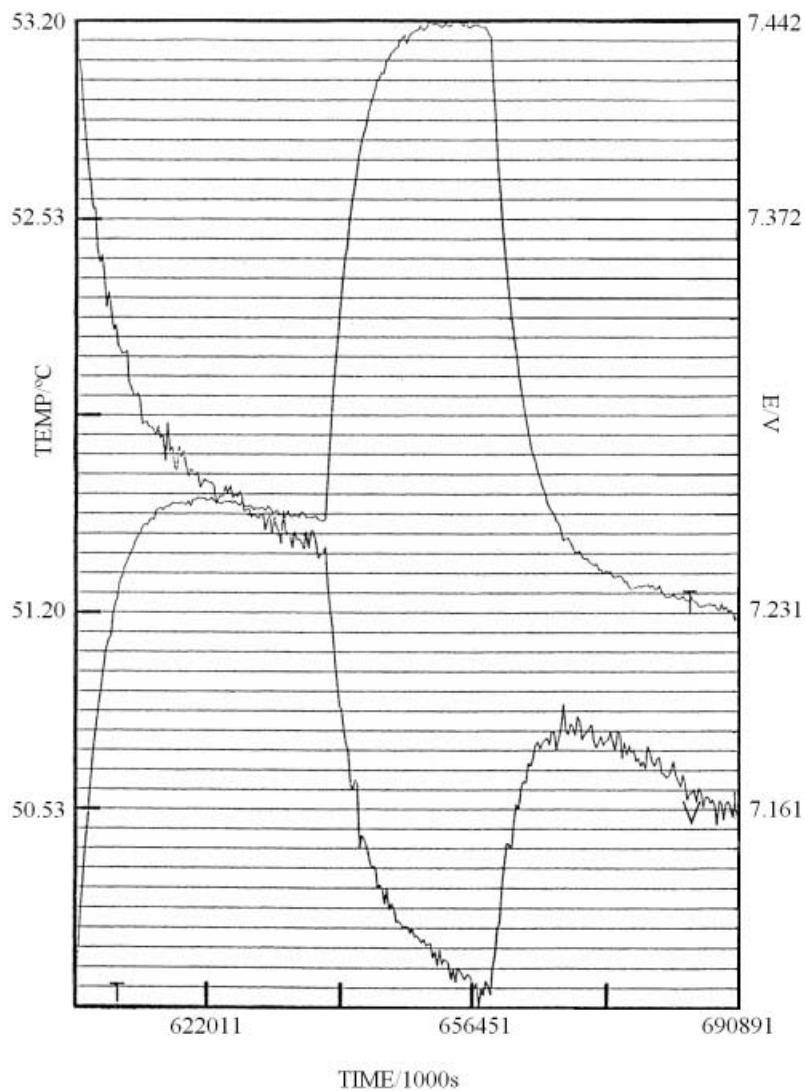


Fig. 5A. calibration of an Icarus cell during the early stages of experiment 4661 conducted by N.H.E. Cathode: 0.4cm diameter by 1.25cm length polarised in 0.1M LiOD/D₂O; cell current 0.5A; calibration pulse $\Delta Q = 0.2504W$

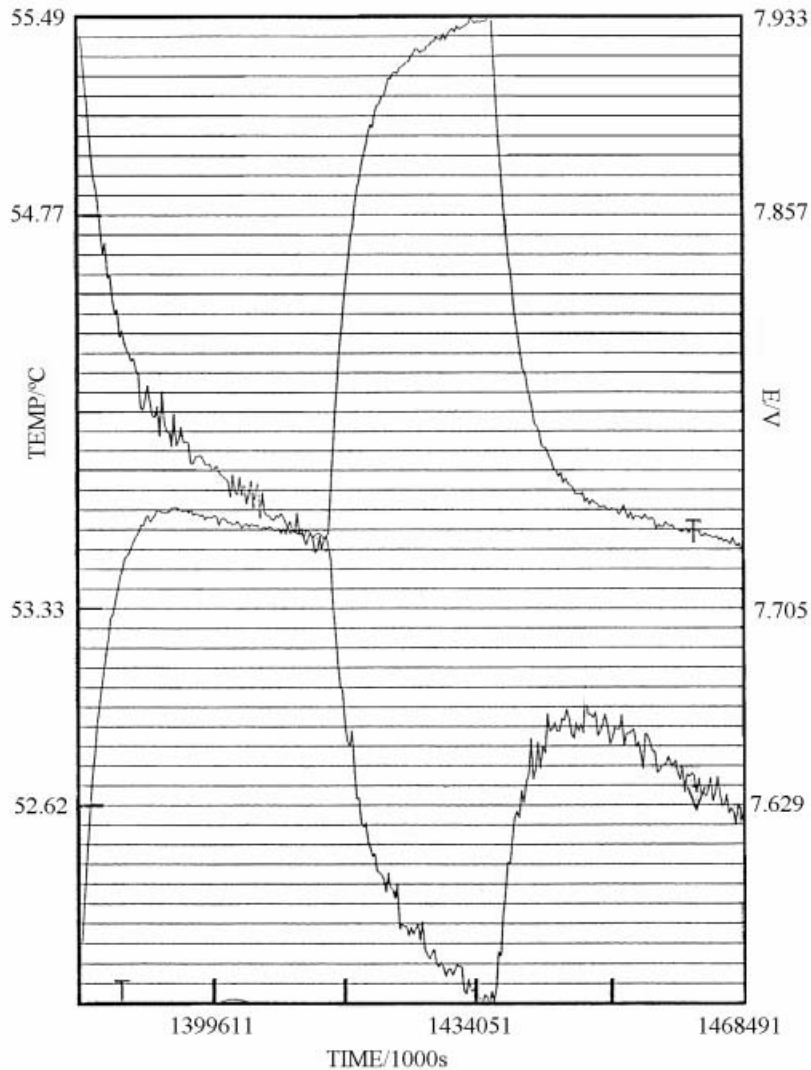


Fig. 5B. A calibration during the later stages of experiment 4661

We must also acknowledge the fact that in the presence of excess enthalpy generation a part of this excess enthalpy may be generated outside certain designs of calorimetric cells (e.g. by the intervention of soft x-rays). This is a further factor which could lead to invalid estimates of the accuracy and precision of calorimeters investigated solely by using the Pd-based systems in D_2O -based electrolytes.

Our Conference Chairman has suggested that it would be useful if I were to give an indication of the papers on the general area of "Cold Fusion" which I have found especially interesting. I have found this to be an invidious task because, having Catholic tastes in science, I have found all the papers interesting even when I have approached these with some scepticism. Moreover, the field of publications is now very wide. However, Table 1 gives some indication of those which have aroused my special interest.

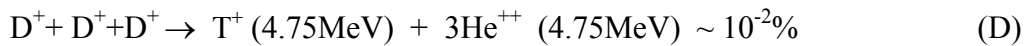
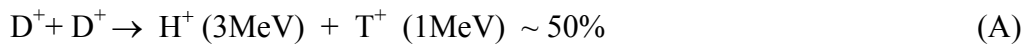
TABLE 1 SOME MILESTONES IN COLD FUSION RESEARCH

G. Preparata et al	Theory of the Subject, Chapter 8 in (1), (16).
P. Hagelstein	Theories of the Subject, (17).
Scott R. Chubb	Theories of the Subject, (18)
Y. Kim	Theories of the Subject, (19)
E. Del Giudice et al	Theory of transmutations at lower energies, (20).
F.	
Y. Iwamura	Low energy transmutations, (21).
G.H. Miley	Low energy Transmutations, (22).
M.H. Miles	Correlation of excess heat with production of ^4He (23); fluidized beds (26).
M. McKubre	Correlation of excess heat with production of ^4He; flow calorimetry. (24)
J.O'M Bockris	Generation of ^4He in the lattice; transmutations (25)
E. Del Giudice F. De Ninno et al	The importance of the vector potential; generation of ^4He; melting of electrodes, (29), (30).
F.E. Cecil et al and others	Charged particle emissions, (27)
D. Letts	Laser stimulation (28).
E. Takahashi	“Hot Fusion” processes at low incident Deuteron beam energies.
Y. Arata	Generation of heat in small particles (35); see also comments in main text (37)
S. Szpak	Hot spots and mini explosions (38); codeposition of Pd and D, (39), (40).
G. Mengoli	Heat after Death (after effect), (41).
Dispersed authorship	isotopic abundances; evidence for photofission; transmutations.

I note in the first place, the papers which attempt to find a theoretical basis for the subject, Chapter 8 in (1), (16)-(19). Clearly, it will prove to be impossible to advance the research in the absence of such a theoretical foundation (or, at any rate, progress would be greatly impeded). As my interest in the subject area was part of a general investigation of the rôle of Q.E.D. in the Natural Sciences, I have naturally been strongly drawn to the approach of Giuliano Preparata.

It will be seen that I have separated from this section (dealing with theoretical aspects) the paper by Del Giudice, De Ninno and Frattolillo covering a possible consequence of Q.E.D. Coherence namely, the question of whether we can explain transmutations at low energies (20). This subject area (21) (and see also (22)) originally seemed quite unreasonable. It also remains to be established how the generation of ^4He (23) (24) is related to the wider area of low energy transmutations. The contributions of Miles and McKubre are noteworthy in that they establish a correlation between excess heat production and the formation of ^4He ; that of Bockris and his co-workers in that they establish the formation of ^4He in the lattice of Pd. I note also that Miles has reported the first measurements using fluidized beds of Pd particles (26). The work of McKubre et al using flow calorimetry has established that excess heat production is not an artefact of isoperibolic calorimetry !

The listing in Table 1 has also singled out a number of other contributions including that on charged particle emissions (27) and laser stimulation of excess heat production (28). Measurements at much lower incident energies of the deuteron beam than those used hitherto in the investigation of "Hot Fusion" have shown that more complicated collision processes come into play at such lower energies (such as the channel (D) (29) than the channels (A) - (C) which have been observed in "Hot Fusion" :



One can therefore speculate whether channel (C) which has a very low cross-section for fusion at low pressures, may not have a much higher cross-section for fusion in a lattice where the production of ^4He is evidently not accompanied by the generation of a γ -ray (1)

The development of the theoretical studies (1), (16) has led to a recognition that one can use the electric potential developed along a thin strip (a component of the vector potential) to enhance the generation of excess enthalpy (30), (31). This is in fact the Quantum Mechanical consequence of coherence in the electrodiffusion of deuterium in palladium.¹³ The formation of ^4He commensurate with excess enthalpy generation has been observed. Under extreme

¹³ The electrodiffusion of hydrogen in palladium was first observed by A. Coehn in 1929 (32). The propagation of the γ -phase of Pd-H along a wire is discussed in (33).

conditions the thin strips melt (boil ?) in the most negatively polarised region.¹⁴ In this context, I also note the work of Arata (36) on the generation of excess enthalpy in nanoparticles of palladium. A plausible interpretation of these experiments is that these particles are destroyed by a single step (or, at any rate, by a small number of steps).¹⁵ Our own consideration of such limiting patterns of behaviour (37) led us to the consideration of the compound Na_2ReD_9 . At the time of our discussions, this compound did not exist whereas Na_2ReH_9 was known. Moreover, two research groups expert in the Chemistry of relevant syntheses, failed to make the deuterated species. Although it is possible to devise numerous arguments to explain the non-existence of Na_2ReD_9 , the simplest of these is that this compound disintegrates on the typical time-scales of chemical syntheses. It is evident that we need to develop “calorimetric syntheses” to study the production of such potentially high energy materials.

In this context, we should also note the direct thermal imaging of “hot spots” on the surfaces of electrodes using infra-red imaging (38). Developments of this methodology (e.g. by using scanning laser-thermometry) should allow the direct determination of the space-time distributions of the fusion steps as well as of the Q-values of these steps. Two of the authors have also introduced a new variant of the electrochemical method of generating excess enthalpy the electrolytic codeposition of palladium and deuterium (39). It has been shown that this method leads to enhanced rates of excess enthalpy generation as compared to the standard method of electrolytic charging of massive electrodes with deuterium (40).

I note finally an extreme example of enthalpy generation but at zero enthalpy input at temperatures close to the boiling point of the electrolyte, a phenomenon which has been variously called “After-Effect” (41), “Heat-after-Death” and “Heat-after-Life”. Such enthalpy production has been maintained for durations of up to 8 days.

I believe that the work carried out thus far amply illustrates that there is a new and richly varied field of research waiting to be explored. Moreover, it seems likely that it will be possible to develop new sources of energy which will be able to operate over a wide range of conditions. Most of the work to date has been carried out at temperatures below the boiling point of the electrolyte but we note that more than 50% of the worlds energy is consumed at temperatures below 70°C. We also note that it would be relatively straightforward to raise the Quality of the heat by using well-established methods.

¹⁴ The importance of this observation is that we can estimate the specific rate of excess enthalpy generation required to achieve this condition. These estimates lie in the range $0.5\text{-}50\text{MWcm}^{-3}$ depending on the nature of the assumptions. Such high rates must be contrasted with the prolonged generation of excess enthalpy at the boiling points of the electrolyte in a “conventional” electrochemical system where the specific excess rate remained restricted to $\sim 2\text{KWcm}^{-3}$ (34), (35).

¹⁵ Which will explain our choice of “massive” electrodes for our investigations.

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