COLD FUSION; PAST, PRESENT & FUTURE.

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In reviewing first of all some of the early history of research into the nuclear reactions of D + compressed into host lattices we need to consider the applicability (or otherwise) of existing paradigms:

Paradigm	Feasibility of Cold Fusion?
Classical Mechanics, C.M.	Question not relevant.
Quantum Mechanics, Q.M.	Not feasible without special assumptions.
Quantum Field Theory, Q. F. T.	Feasible.

Here, we should reserve our judgement: it might turn out, for example, that some special assumptions are justified. However, it is best to observe initially the principles of "Occum's Razor" and this points us to interpretations in terms of Q.F.T. The key question we need to pose is: do the ground states defined by the many body problem make the nuclear states accessible at low temperatures? Here, we have to admit that the application of Q.F.T. to condensed matter (for a summary see (1)) is not generally understood (readers may find the formulation in terms of the Hamiltonian (2) more accessible than that in terms of the Lagrangian coupled to the use of path integrals (1)). It follows, therefore that those relying on Q.M. will judge research in this field to be mistaken (especially if they will not countenance special assumptions!)

Our initiation of research in this field was strongly influenced by our conclusion in the late 1960's that the behaviour of H+ and D+ electrochemically compressed into Pd-host lattices (3) could only be understood in terms of Q.F.T. This led us in 1983 to pose the following two questions:

(i)would the nuclear reactions of D+ compressed into host lattices be different to the reactions in a dilute plasma?

(ii)could such effects be observed?

We expected the answer to (i) to be "Yes" and the answer to (ii) to be "No". Nevertheless, we started a limited investigation and considered experiments based on the following options:

A. Electrodiffusion;

B. Electrochemical charging; plasma excitation;

C. Highly reducing / superbasic media;

D. Highly oxidising / superacid media; the link to "Hot Fusion";

Although our primary objective was A, we started with B (as a preliminary to A) because we believed that such systems are closest to the dictates of Q.F.T. Discussion of the possible influence of plasma excitation is beyond the scope of the present report although it should be noted that fragmentory evidence points to its possible importance. It should be noted though that consideration of this aspect led to the topic now known as Surface Enhanced Raman Spectroscopy, SERS.

As is well-known, we opted initially for calorimetry as the primary "catch-all" methodology. \ Moreover, the calorimetric method chosen had to meet a number of important criteria which included: conformation to "ideal behaviour" (implying predictability from the laws of Physics; we opted for the "well-stirred tank" familiar from the field of Chemical Engineering); high stability of the thermal impedances; uniformity of the temperature throughout the volumes of the cells; possibility of non-isothermal operation; high precision and accuracy and, needless to say, low unit cost. These criteria dictated the choice of isoperibolic calorimetry.

As is well-known the results obtained have been very surprising and include: generation of excess enthalpy far beyond the limits of any conceivable chemical processes and not accompanied by the commensurate generation of tritium and neutrons predicted from measurements of "Hot Fusion" (4), (5), the onset of "positive feedback" at prolonged times and elevated temperatures (i.e. the increase of excess enthalpy generation with increases of temperature e.g. see (6)); "Heat after Death" (in the most extreme form generation of enthalpy at near zero enthalpy input at temperatures close to the boiling points (7)).

Evidently such extravagant conclusions require careful substantiation. A necessary first step is the investigation of suitable "blank experiments" to determine the limits to which zero excess enthalpy can be measured by any chosen calorimetric technique. For the case of isoperibolic calorimetry we observe that all such measurements are entirely normal. Thus Fig 1 is an illustration of a measurement cycle for an experiment using a Pt-cathode. Normality throughout the duration of the experiment is shown by three principal signatures: first of all the heat transfer coefficients vary in an entirely sensible way with time, Fig 2, the decreases being due to the lowering of the electrolyte level due to progressive electrolysis; secondly, the relaxations of the temperature, Fig 3, can be predicted in all regimes by an approximate analysis allowing for the normal "negative feedback", the lowering of the enthalpy input due to increases of the temperature, Fig 1 (see Appendix 4 of (5)); thirdly, the "point-by-point" rates of excess enthalpy generation are normally distributed, Fig 4. For the particular example illustrated, the mean rate of excess enthalpy generation is 79 μW with a standard deviation of 5.87 mW. However, this standard deviation is <u>not</u> an estimate of the accuracy because the overall thermal balance is determined by aggregating the individual values, here 2200 measurements. This aggregate, Fig 5, necessarily agrees with the mean, Fig 4, and this methodology shows that it is more appropriate to use the integrals of all the variables in the first place. This is the basis of the ICARUS methodology (8) based on the analyses developed in 1992 (9).

The values of the excess enthalpies determined in "blank experiments" of this kind are about one order of magnitude less than those for the reduction of oxygen

present at the equilibrium concentration (partial pressure 1/3 atmosphere). The reason for such low values is simple: the stream of cathodically evolved deuterium degasses the oxygen in the boundary layer. Repeated explanations of excess enthalpy generation in terms of such cathodic reduction are unfounded (note also that such explanations are excluded by the material balances which have been repeatedly measured viz. the volumes of the evolved gases and the D_2O additions required to maintain the electrolyte levels as determined by Faraday's Laws of Electrolysis).

Results such as those shown in Figs 4 and 5 are obtained using heat transfer coefficients determined by repeated calibrations (a topic beyond the scope of the present article). The performance is somewhat degraded if a single calibration is used in view of irreproducibility in replenishing the cells with D_2O. However, the primary reason for the repeated calibration of the cells is the monitoring of the system behaviour. We note that whereas "blank experiments" are always entirely normal (e.g. See Figs 1 - 5) it is frequently impossible to find any measurement cycle for the Pd-D_2O system which shows such normal behaviour. Of course, in the absence of adequate "blank experiments" such abnormalities have been attributed to malfunctions of the calorimetry, e.g. see (10). However, the correct functioning of "blank experiments" shows that the abnormalities must be due to fluctuating sources of excess enthalpy. The statements made in this paragraph are naturally subject to the restriction that a "satisfactory electrode material" be used i.e. a material intrinsically capable of producing excess enthalpy generation and which maintains its structural integrity throughout the experiment. Most of our own investigations have been carried out with a material which we have described as Johnson Matthey Material Type A. This material is prepared by melting under a blanket gas of cracked ammonia (or else its synthetic equivalent) the concentrations of five key classes of impurities being controlled. Electrodes are then produced by a succession of steps of square rolling, round rolling and, finally, drawing with appropriate annealing steps in the production cycle.

An illustration of the complications observed for measurements on the Pd-D_2O system is given in Fig 6; this is a portion of the temperature and cell potential-time series for the second experiment described in (9). It is found that with increasing time and temperature the relaxations can no longer be predicted, the deviations requiring the superposition of an endothermic absorption process on the normal behaviour, Fig 7 (compare Fig 3 and see also (6)). In turn, this additional absorption leads to an increase in the rates of excess enthalpy generation so that the temperature rises due to the calibration pulse are larger than predicted, Fig.6. It goes without saying that calibration of the cells is impossible under such conditions: in particular, the "lower bound heat transfer coefficients" (based on the assumption of zero rates of excess enthalpy generation) are found to be larger than the "true heat transfer coefficients", a result which contravenes the Second Law of Thermodynamics! (this has been pointed out repeatedly e.g. see (6)).

With the evident impossibility of "on-line" calibrations of the cells, we need to make conservative assumptions in evaluating the excess enthalpy generation. For example, we can use calibrations made before the onset of "positive feedback" and ensure that the cells can never operate endothermically, compare Figs 8 and 9. We note that even if we make an incorrect choice of the heat transfer coefficients so

that the cell is initially endothermic (forbidden by the Second Law of Thermodynamics), we nevertheless still observe excess heat generation beyond the reach of any chemical explanation even for a single day's operation, Fig 9 (excess enthalpy per mole of Pd or LiOD.)

The development of "positive feedback" is of crucial importance to the social significance of the topic because it shows that elevated levels of energy production should be feasible at least at temperatures adequate for the generation of "low grade heat" (say at temperatures up the boiling points of the electrolytes). Investigation of energy production under such conditions requires modifications of the calorimeter designs. One approach is to use the ICARUS 9 calorimeter (11) in which a lower section mimics the behaviour of the ICARUS 1 type while an upper section allows the measurement of heat flows at the boiling points. Fig 10 illustrates a measurement on a Johnson Matthey Material Type A carried out in 1994. Such measurements have shown that it is possible to achieve sustained high levels of excess enthalpy generation (1-2 $kWcm^{-3}$) at an excess of 100% of the power input for prolonged periods (~ 50 days).

An alternative approach is to use an auxiliary stream of inert gas (N_2) to control the rate of evaporative cooling of calorimeters operating close to the boiling point (12). Adjustment of the rate of gas flow then gives control of the calorimeter constant. These measurements have shown that there is no threshold current density for the onset of excess enthalpy generation above the onset of "positive feedback", Fig 11 whereas there is such a threshold below this point (5). It is evidently now of crucial importance to determine the parameter spaces at the very least before and after the onset of "positive feedback."

There is, however, a further twist to this topic because it is found that following operation of the cells at the boiling points (9), (7), or close to this temperature (12) there is enthalpy generation even at very low or even zero enthalpy input, e.g. see Fig. 12. This phenomenon, which is now variously called "Heat after Death", "Heat after Life" or "After Effect" requires intensive investigation so as to define the limits of the various categories under which it might be observed and (7) to integrate the effects with operation of the cells using pulsed currents (so as to achieve energy efficient systems at high power densities). The work so far carried out shows that it should be possible to develop demonstration devices at an early date and having specific thermal outputs in the range 5 - 250Wcm^{-3} (based on the volume of the electrodes). At the low to middle part of this range (say 5-50Wcm^{-3}) such devices would only find applications in niche markets; at the upper end of the range the devices could find very extensive applications with foreseeable increases in the world production of Pd (say, by a factor of 10).

It is apparent, however, that the further development topic requires the investigation of systems based on electrodiffusion, A above, both from the point of view of the applications of the research as well as for the investigation of the basic science. The feasibility of using such an approach is foreshadowed by the work of Coehn who showed that hydrogen is present as protons in the lattice (13) (for a discussion of the background to this topic see (14). Consideration within the framework of Q.M. of the applicability of this approach for creating highly loaded host lattices leads to the model illustrated in Fig. 13. However, within the framework of Q.F.T. (15) and, by analogy to the Bohm-Aharonov Effect (for a summary of research on this effect up to 1989 see (16)) which Preparata describes

with the epithet "Coehn-Aharonov Effect", we arrive at the model shown in Fig 14. The electrochemical potential of D+ in the lattice is now controlled by differences in the Galvani potential rather than by gradients of this quantity thereby allowing the creation of much more extreme conditions than can be achieved by electrochemical or chemical charging alone. Furthermore, such an approach leads to confinement of D+ within the lattice. It has been shown that excess enthalpy generation at the level $10kWcm^{-3}$ can be achieved in thin wires in this way (and outputs up to ~ $100kWcm^{-3}$ have been demonstrated (15)). If we focus attention once again on the social significance of such results we observe that enthalpy generation at such levels would permit the replacement of most existing energy producing systems with world-wide production of Pd at the existing levels. The scientific importance lies in the fact that whereas the Bohm-Aharonov Effect is a clear demonstration of the need to replace the C.M. by the Q.M. paradigm, the Coehn-Aharonov Effect (indeed, "Cold Fusion" in general) is a demonstration of the need to go one step further to the Q.F.T. paradigm.

Some further observations.

It will be observed that notwithstanding the wide range of the title, this report has taken a very narrow "cut" through the published information (much narrower indeed than that outlined in the Abstract). The major reason for focusing attention on the particular aspects outlined here is my belief that acceptance of the topic still awaits the construction of applicable demonstration devices. Furthermore, restrictions on the length of this report requires the exclusion of most of the published information and such exclusion can by no means be taken as a value judgement!

The setting of an early technological target (which I believe has been a common feature of much of the research in this field) has created great difficulties because the knowledge base is far from complete. Thus, as far as the major signature, the generation of excess enthalpy, is concerned we still need to establish the role of the materials variables, the parameter spaces as a function of temperature, the role of changes in the experimental protocols, the condition required for energy efficient operation (especially the effects of pulse operation), the effects of choice of system (e.g. the effects of changing from B to A or C, the effects of using C + A or B + A), the special requirements posed by the construction of demonstration devices and much else! Furthermore, it is necessary to integrate all the observations (tritium, neutrons, energetic particles, creation of elements with "unnatural" isotopic abundances, emission of radiation), into a general scheme. We need to ask: are these observations due to transients observed during the establishment of extensive fully coherent states or are they correlated with the major signature? what is the role of photonuclear reactions? will the observations of "Hot Fusion" at low particle energies (perhaps better described as "Warm Fusion" e.g. see (17)) converge onto those of "Cold Fusion"? can we arrive at an interpretation in terms of Q.F.T. of the various interesting signatures which have been obtained in studies of the application of Mechanical Forces on Lattices i.e. of C.M. (e.g. the topic of Structure Breaking Energy Release, S.B.E.R., whose disclosure and discussion is long overdue)? and what can we learn from the application of Q.F.T. to more conventional problems in the Natural Sciences?

The results obtained to date lead to a set of questions which in many respects are more interesting than the results themselves.

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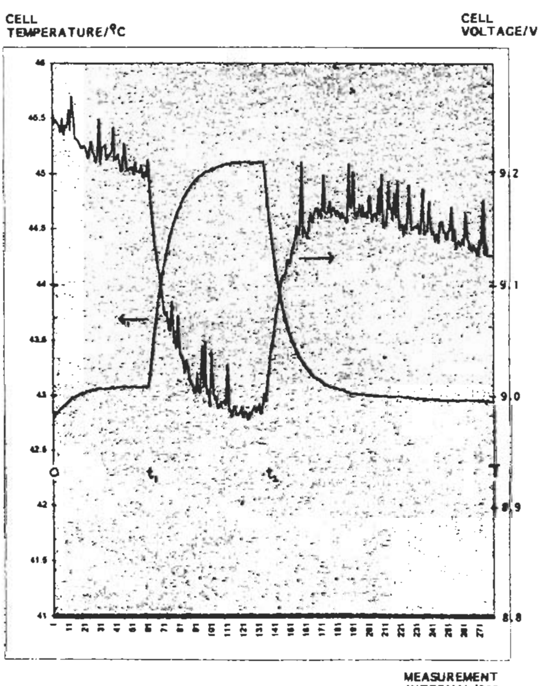

Fig. 1 The cell voltage- and cell potential - time series for the 14th day of polarisation of a Pt cathode in 0.1M $LiOD/D_2O$; Cell current : 0.25A.

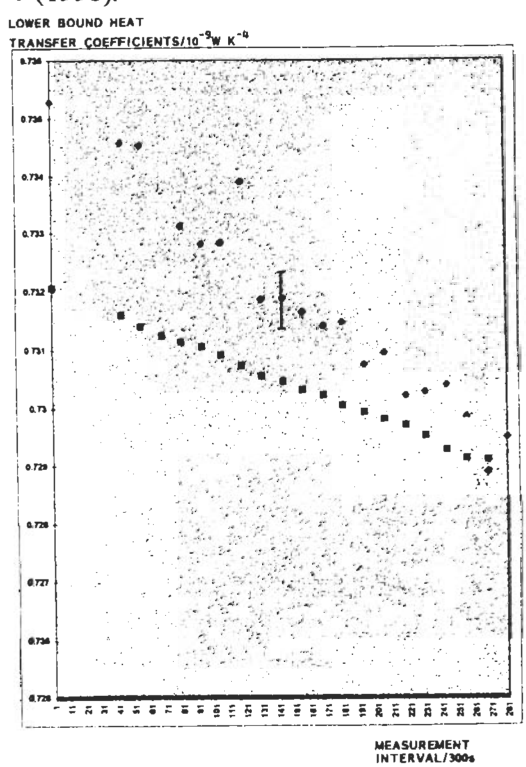

Fig. 2 The lower bound heat transfer coefficients for the first day of polarisation of a Pt cathode in 0.1M $LiOD/D_2O$, Cell current: 0.25A

—◆— $(k_R')_{11}$, the 11-point mean of the "point-by-point" (differential) measurements; I denotes $\pm\sigma$ of the values.

—■— $(\overline{k_R'})_{21}$, the integral values based on the backward integration of the data sets

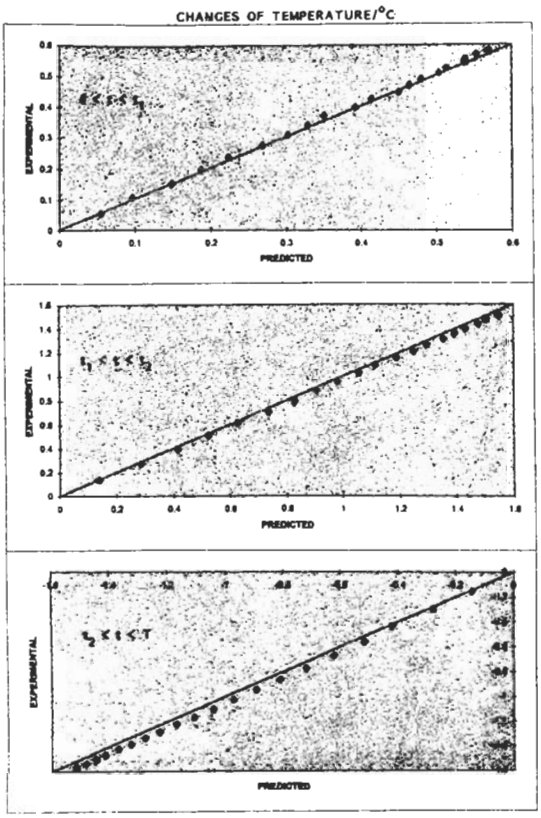

Fig. 3 The experimentally observed temperature relaxations plotted against the predicted values for the time regions $0 < t < t_1$, $t_1 < t < t_2$ and $t_2 < t < T$ of Fig 1. The predicted values take into account the "negative feedback" shown in Fig. 1.

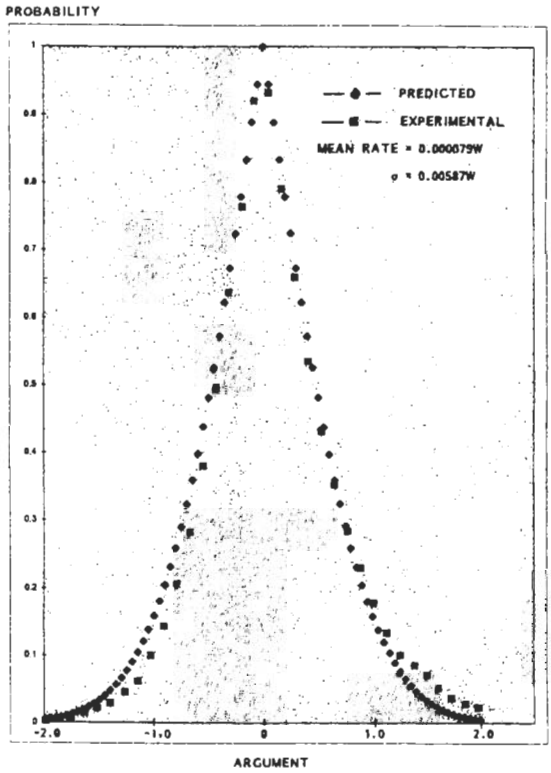

Fig. 4 The upper and lower tail distributions of the observed "point-by-point" excess enthalpies in the time region $t_2 < t < T$ for 20 measurement cycles of a Pt cathode polarised in 0.1MLiOD / D_2O; cell current : 0.25A.
Cell recalibrated for each measurement cycle.

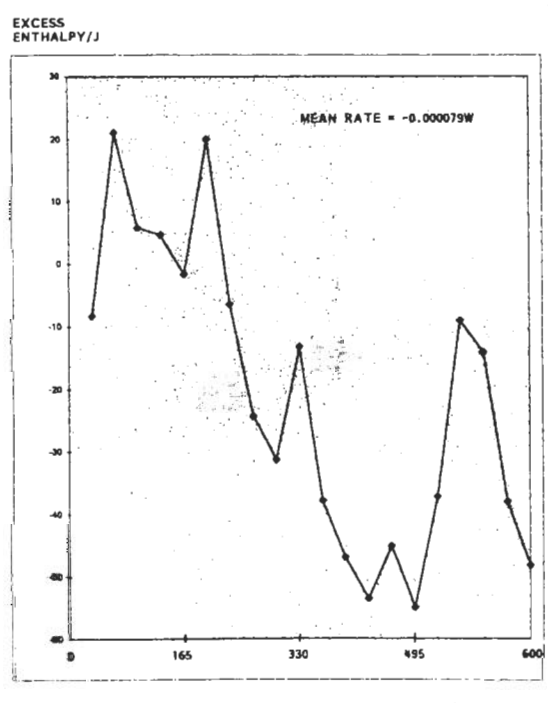

Fig. 5 The total excess enthalpy as a function of time for the data sets illustrated in Fig. 4. Cell recalibrated for each measurement cycle.

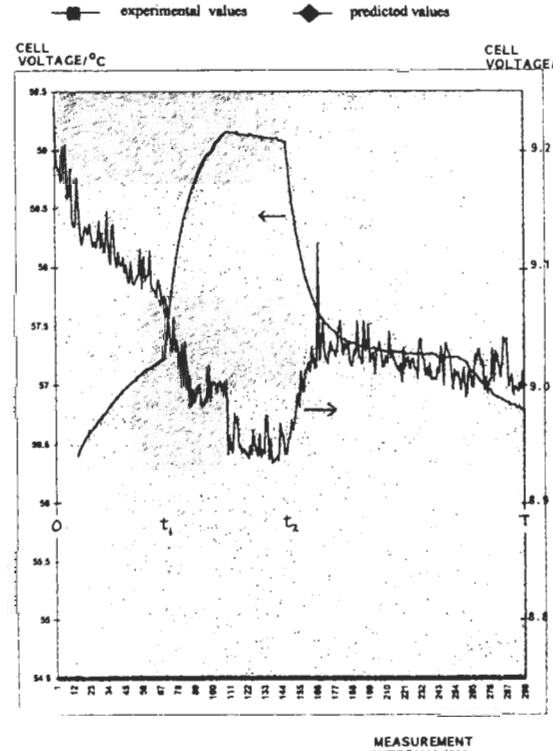

Fig. 6 The cell voltage - and cell potential - time series for the 6th day of polarisation of a Pd rod cathode (ϕ = 0.2cm, ℓ = 1.25cm) in 0.1M LiOD/D_2O; cell current: 0.5A.

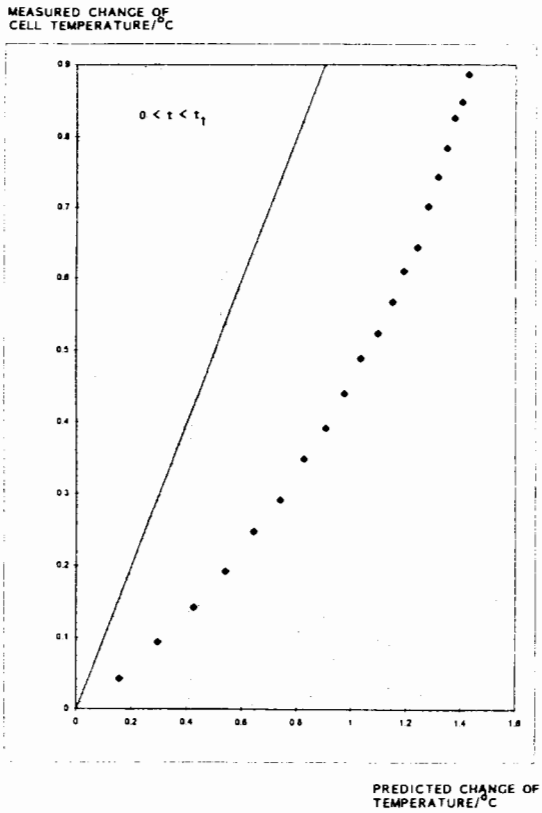

Fig. 7. The experimentally observed temperature relaxation plotted against the predicted values for the region $0 < t < t_1$ of Fig. 6. The predicted values take into account the "negative feedback" shown in Fig. 6.

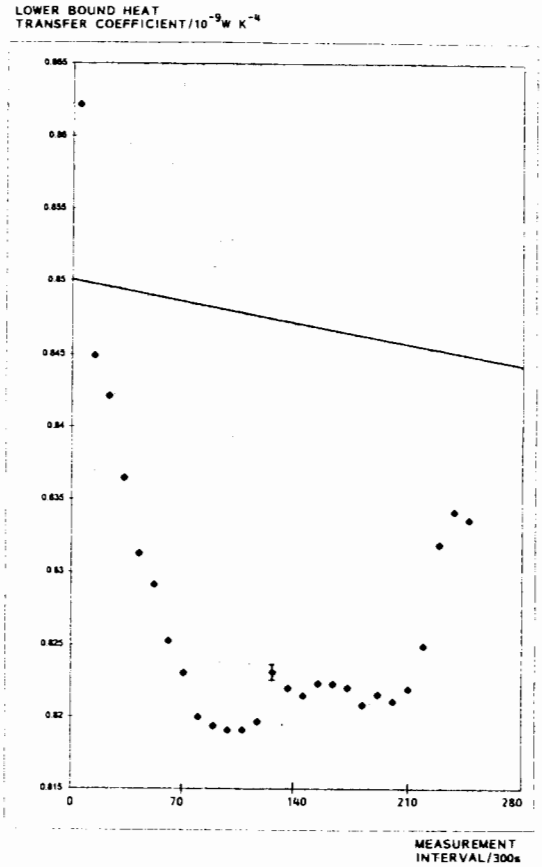

Fig. 8. The lower bound heat transfer coefficient $(\overline{k_R^{-1}})_B$ as a function of time for the data set shown in Fig. 6. The full line shows an incorrect low estimate of the true heat transfer coefficient $(\overline{k_R^{-1}})_M$. I denotes $\pm\sigma$ of $(\overline{k_R^{-1}})_M$.

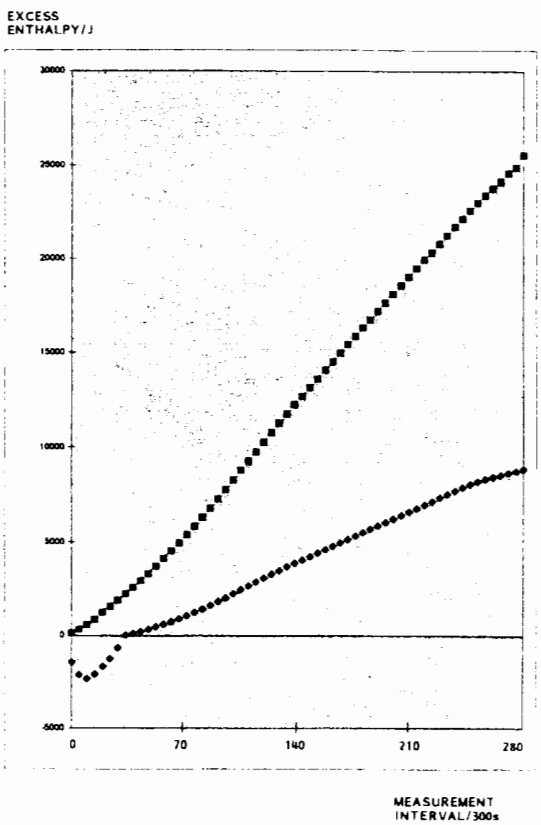

Fig. 9. The total excess enthalpy for the data set illustrated in Fig. 6.

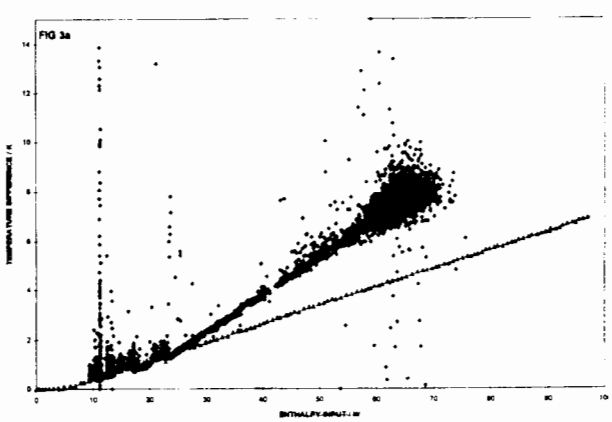

Fig. 10. Prolonged measurements in an ICARUS 9 calorimeter of excess enthalpy generation in a Pd cathode (ϕ = 0.2cm, ℓ = 1.25 cm, Johnson Matthey Material Type A).

calibration of the upper reflux section

experimental measurements

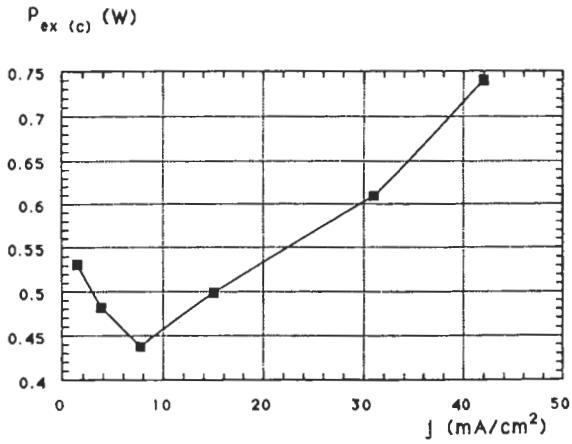

Fig. 11. The rate of excess enthalpy generation as a function of current density at temperatures close to the boiling point. Pd-sheet cathode (0.02 x 1.3 x 2.5 cm.) polarised in 0.6M K_2CO_3 /D_2O.

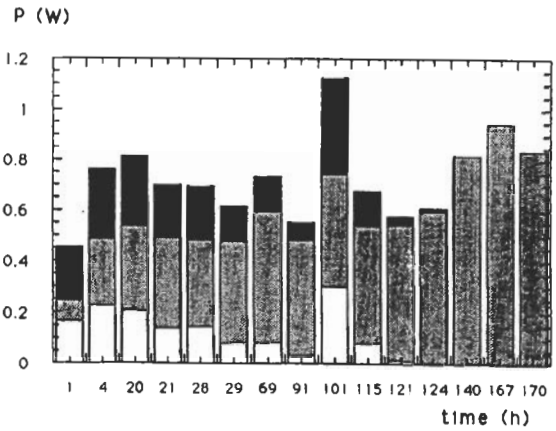

Fig. 12. The rate of excess enthalpy generation as a function of time at temperatures close to the boiling point. Pd-sheet cathode (0.02 x 1.3 x 2.5cm) polarised in 0.6M K_2CO_3 / D_2O.

□ Enthalpy input, ▨ enthalpy output, ▩ correction for evaporation into the electrolytically generated gas stream.

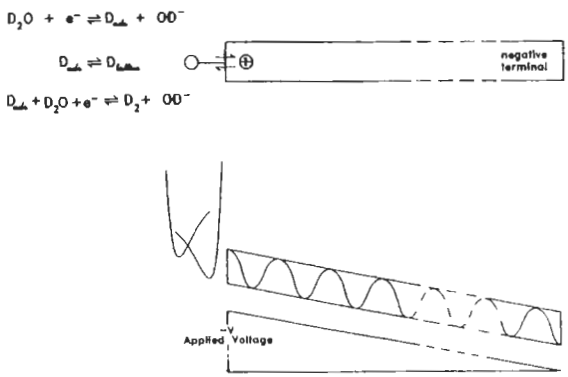

Fig. 13. Electrodiffusion in a Pd-wire as predicted by C.M. + Q.M.

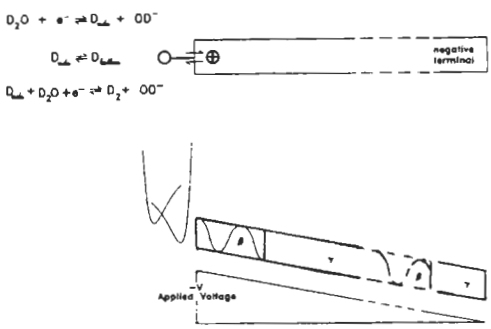

Fig. 14. Electrodiffusion in a Pd-wire as predicted by Q.F.T.