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Mol, September 25, 1989

Concerns : "Cold fusion"

Ref. : F8319092

Dear Sir,

Please find enclosed a report concerning our observations of excess heat production during so-called "cold fusion" experiments and an explanation for the phenomenon we have observed.

We have sent this report to the laboratories where excess heat production has been observed as well, with the request to investigate whether their observations might be explained in a similar way.

We would be grateful if you could send us your comments.

Yours sincerely,

M. SNYKERS

Programme Manager
Fusion Technology

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STUDIECENTRUM VOOR KERNENERGIE

FUSION TECHNOLOGY

CENTRE D' ETUDE DE L' ENERGIE NUCLEAIRE

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EXPERIMENTAL EVIDENCE OF ERRONEOUS HEAT PRODUCTION IN COLD FUSION EXPERIMENTS

A. Bruggeman, M. Loos, C. Van der Poorten, R. Craps,
R. Leysen, F. Poortmans, G. Verstappen, M. Snykers

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ABSTRACT

In the course of an electrochemical loading "Fleischmann and Pons type" experiment, an excess heat production has been observed in the cold fusion laboratory of the Nuclear Research Centre in Mol, Belgium. This excess heat was not due to fusion reactions and could not be explained on physico-chemical grounds.

An explanation is given on the basis of a hidden input of electrical energy from the power source, which is not revealed by the standard DC measurements. It consists of a low frequency AC current which is superposed on the applied DC current and may be generated by the occurrence of an oscillation instability in the electrical circuit of the cell.

The actual excess heat production phenomenon could be reproduced during simulation experiments.

1. Notes on wording: "an explanation is given." This language goes beyond their findings. See their conclusion. More accurate language based on their findings is "a possible explanation is given."
2. There are two components to the excess heat production phenomenon in this paper. The 30-minute steep rise (which the authors address) and the 2-hour out-of-control thermal runaway (which the authors mention but do not address).
3. That the authors could, in simulation experiments, impose a hidden AC current to amplify the measured power, does not at all mean that that is what happened to these cells.
4. In the real experiment, the decrease in voltage, concurrent with the increase in cell temperature, provides direct physical evidence of a) cell heating concurrent with b) reduced measured input power.

1. INTRODUCTION

In the search of cold fusion based on electrochemical loading of palladium with deuterium, several research teams have observed an as yet unexplained excess heat production [Fleischmann and Pons [1], Appleby [2], Huggins [3], , Kainthla [4] and Gozzi (Univ. Rome)].
Attempts to explain this heat production were mainly based on electrochemical or physico-chemical grounds. They were not conclusive [5] [6]. Soon after the Fleischmann and Pons observations became known to the public, similar electrochemical experiments were started at the Nuclear Research Centre S.C.K./C.E.N. The main aim of the programme was to investigate the origin of the excess heat production. Particular attention was paid to the possible influence of various electrochemical parameters.

2. EXPERIMENTAL SET-UP

The experiments have been performed in a specially equipped laboratory, with two cells running in parallel, one with LiOD-D_2O and one with LiOH-H_2O.
The heart of each set-up is a glass electrolysis cell with an outer diameter of 4.5 cm as shown by Fig. 1. As electrolyte solution 0.1 M LiOD in D_2O respectively 0.1 M LiOH in H_2O is used, which is similar to the one used by Fleischmann and Pons. The level in the cell is kept constant by regular addition of D_2O respectively H_2O, except after sampling when 0.1 M LiOD in D_2O respectively 0.1 M LiOH is added. For both cells, the quantity of electrolyte solution is about 50 ml.

The electrolysis cells are equipped with 3 electrodes :

- A central palladium rod (Pd), or working electrode, which may be loaded cathodically with D (H). The Pd electrodes used are cylinder-shaped rods with a diameter of 0.4 cm and a length of 5.0 cm. At the top they are equipped with a 0.1 cm diameter Pd wire. This wire and also all parts emerging from the solution are electrically insulated. The used enamel constitutes also a diffusion barrier for deuterium (hydrogen).

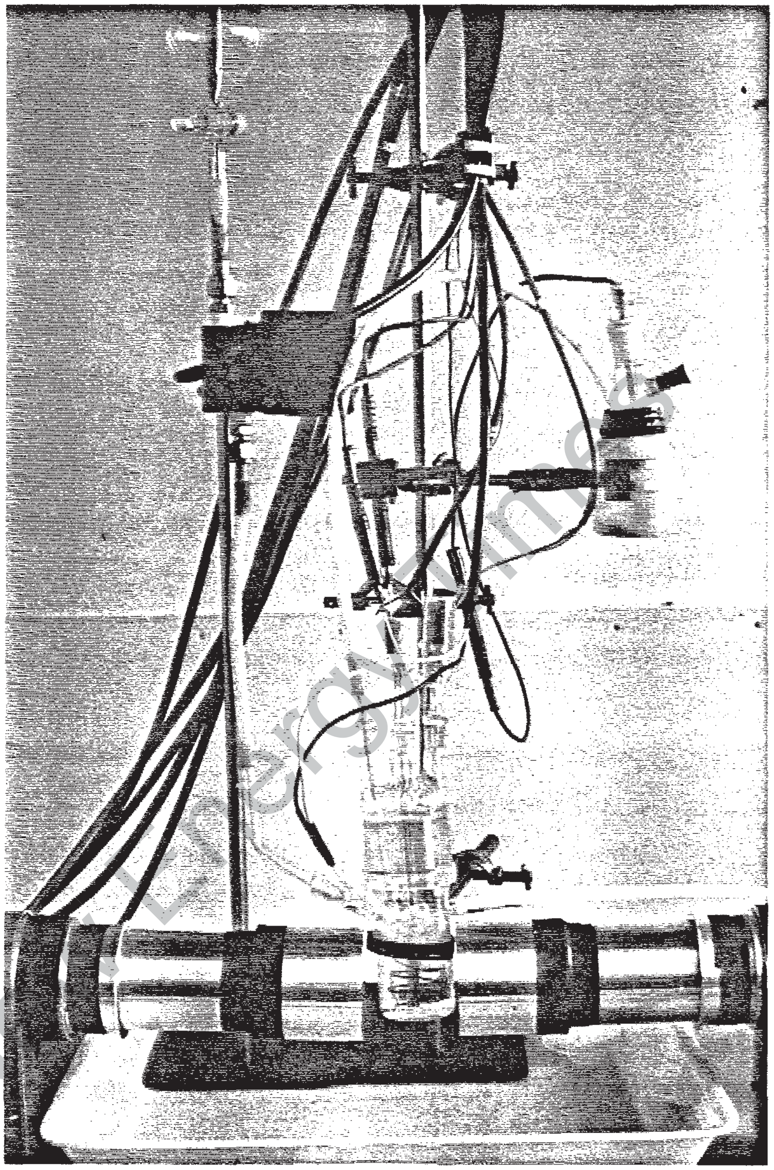


Fig. 1 : Assembly for D_2O electrolysis. In the middle : glass cell with working electrode (Pd), electrolyte bridge to the reference electrode (Hg/HgO) and counter electrode (Pt). Left and right of the cell : liquid scintillation detectors for neutron detection.

To keep the formed D_2 (H_2) more or less separated from the formed O_2, a reversed glass funnel has been placed around the Pd wire and around the upper part of the Pd rod; this funnel is immersed in the liquid and allows the vertically bubbling D_2 (H_2) to escape. The Pd has been supplied by Johnson Matthey and has not been pretreated for the experiments realized up to now.

- A platinum (Pt) electrode or counter-electrode, i.e. an electrode for closing the electrical circuit. This electrode consists of a Pt wire with a diameter of 0.1 cm and a length of about 25 cm; it is wound in a spiral around the Pd electrode at a distance of about 1 cm.

- A Hg/HgO reference electrode, i.e. an electrode for controlling and/or monitoring the electrochemical process. The potential of the Hg/HgO reference electrode at 25°C in 0.1 M LiOH is + 0.166 V with respect to the standard hydrogen electrode. This reference electrode is connected to the electrolyte solution by means of a bridge of LiOD respectively LiOH solution.

A well-determined potential E or current I is obtained by using Amel potentiostats/galvanostats (model 550), and, if necessary, completed by a function generator (Amel, model 567).

Continuous and automatic recording of data is realized by means of a personal computer, a multichannel scanner (SCK type 702), a Schlumberger Solartron voltmeter (model 7060) and the necessary hard- and software.
It concerns the following data.

- The potential difference, E_{ref}, between reference and working electrode, with $E_{ref} = - E_w$, in which E_w is the potential difference of the working electrode compared with the reference electrode (Hg/HgO). The (cathode) potential of the working electrode compared with the standard hydrogen electrode is given by :

$$E_K = - E_{ref} + 0.166 \text{ V} = E_w + 0.166 \text{ V} \tag{3.1}$$

- The total cell tension, U_{cell}, i.e. the terminal voltage between counter- and working electrode.

- The cell current I.
- The temperatures inside (T_{in}) and outside (T_{out}) the electrochemical cell, measured with Pt 100 thermistors not corrected for mutual deviations.

All values are measured about every 15 s. An average value is recorded per adjustable time interval (usually 6 to 8 minutes).

The neutron detection system consists of two independently working proton-recoil liquid scintillators, type NE213 (Ø = 5 cm, h = 5 cm). The pulse-height spectra (2 x 2 K) are stored in a multichannel analyzer with multiplexer. The absolute efficiency of these detectors had been determined previously for various neutron energies between 0.5 MeV and 16 MeV at the Van de Graaff accelerator of CBNM. Pulsed mono-energetic neutron beams were produced with the reactions $T(p,n)^3He$, $D(d,n)^3He$ and $T(d,n)^4He$ and a calibrated proton-recoil telescope was used as reference detector.

The detectors are installed at a few mm from the D_2O cell so that the total efficiency for 2.5 MeV neutrons is 5 % per detector with a bias level at 1.2 MeV. Puls-shape discriminator is used to reduce the background from gamma radiation. The total background per detector is 4.3 10^{-3} counts per second with a bias of 1.2 MeV.

An unshielded 3" x 3" NaI (Tl) detector, coupled to a multichannel analyser with a range of 0.1 to 10 MeV (Canberra series 35 plus), has been installed in the immediate vicinity of the D_2O cell for the measurement of the gamma-rays which might be created directly or by interaction (a.o. in the detector itself) of produced neutrons.

The tritium activity of the used solutions 0.1 M LiOD in D_2O and 0.1 M LiOH in H_2O before a test and the tritium activity in the cells after a test period, are measured by liquid scintillation counting with a precalibrated Packard Tri-Carb device (model 4430) equipped with DPM and Spectraview options. It is supposed that possibly produced tritium finally (partially) arrives in the solution. This hypothesis seems to be justified because of the selective enrichment of lighter hydrogen isotopes in the palladium and of heavier ones in the electrolyte solution [7, 8]. Further details concerning the experimental set-up can be found in ref. [9].

3. EXCESS HEAT PRODUCTION EXPERIMENT

In accordance with the ideas of Fleischmann and Pons [1], the main part of the experiments consisted in a continuous electrochemical loading of palladium electrodes with deuterium respectively hydrogen. Loading was carried out at controlled cathode potential, i.e. at a constant negative potential E_w of the working electrode compared with the Hg/HgO reference electrode. A general survey of experiments performed is given in ref. [9].

The present report only deals with the experiments when excess heat production was observed.

On 16.06.1989, after two months of experimentation, a sudden temperature rise occurred in both cells, as shown in Fig. 2. The two cells were than in operation for 46 h in conditions given in Table 1. The working conditions for both cells had been stable for the past 46 hours.

	I	U_{cell}	$T_{in} - T_{out}$
D_2O cell	0.3 to 0.5 A	4.0 to 4.7 V	3 to 5°C
H_2O cell	0.2 to 0.25 A	3.0 to 3.5 V	1.0 to 1.5°C

E_w = - 1.6 V compared with Hg/HgO
Environmental temperature : 28-30°C

TABLE 1
Working conditions of the cells

The heating of the cells corresponded with a power of more than 20 W. Yet, the measured values of I, U_{cell} and E_{ref} (= - E_w) dropped to lower values than before [Fig. 3]. U_{cell} and E_w were checked at the cell with a portable multimeter and the measured values corresponded

Crucial information here. They acknowledge that the cells went from equilibrium in the previous 46 hours to producing 20W of excess heat in the 30-minute burst beginning at 270h. They noticed the concurrent drop in voltage as expected by E=IR. Their data does not show that current dropped, nor should it have if they were running a FP cell, which runs in contstant-current mode. Including "I" after "measured values of I, Ucell" appears to be an error.

with the recorded (low) values. On the basis of the measured values the dissipated heat had to be less than 0.4 W for both the D_2O and H_2O cell.

After 1 h, when decoupling the function generator, the H_2O cell suddenly normalized. This had no effect on the D_2O cell. Replacement of the potentiostat had also no result. The D_2O system was no longer controllable and the cell had to be cooled by means of a cold air blower. For the D_2O cell, this abnormal situation lasted for about 2 h, so that the dissipation may be estimated at 100 to 200 kJ.

The potentiostats themselves also dissipated more heat than normally. The PdD_n (PdH_n) cathodes behaved rather normally but around the Pt counter electrode of the D_2O cell strings were formed.

After about 2 h, normalization of the D_2O cell suddenly occurred when taking a 5 ml sample from the hot LiOD in D_2O solution in view of the tritium content measurements.

The tritium activity in the D_2O cell was increased by about 65% to 2.6 Bq/g as compared with the initial tritium activity. Because of the known tritium enrichment in the electrolyte solution during electrolysis of D_2O, the measured tritium concentration can not be considered to be abnormally high.

In the course of this process of abnormal heat production, the neutron measurement equipment did not show any evidence of a neutron production, however a previously non observed defect occurred in the gamma measurement circuit. An ordered peak pattern appeared in the channels corresponding normally with energies from 4 to 8 MeV. Above 8 MeV still smaller peaks appeared. This was due to a temporary defect of the measurement equipment.

On the basis of those observations it was clear that the excess heat was not produced by nuclear fusion reactions.
There was also considerable doubt that it was due to a chemical reaction since an energy release of 100 to 200 kJ can be produced by the oxidation of 0.5 ml deuterium, or 10 times the maximum capacity of deuterium to be expected in the Pd D_n cathode.

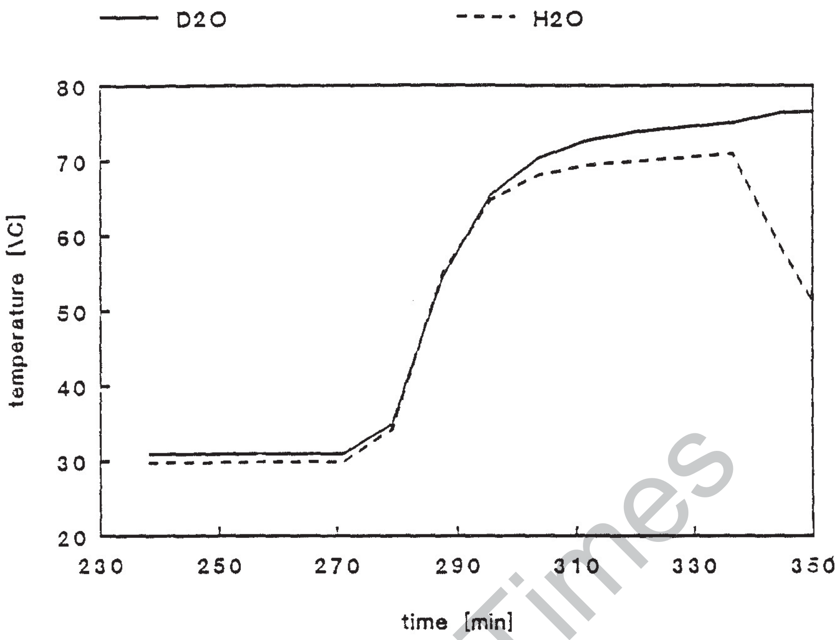
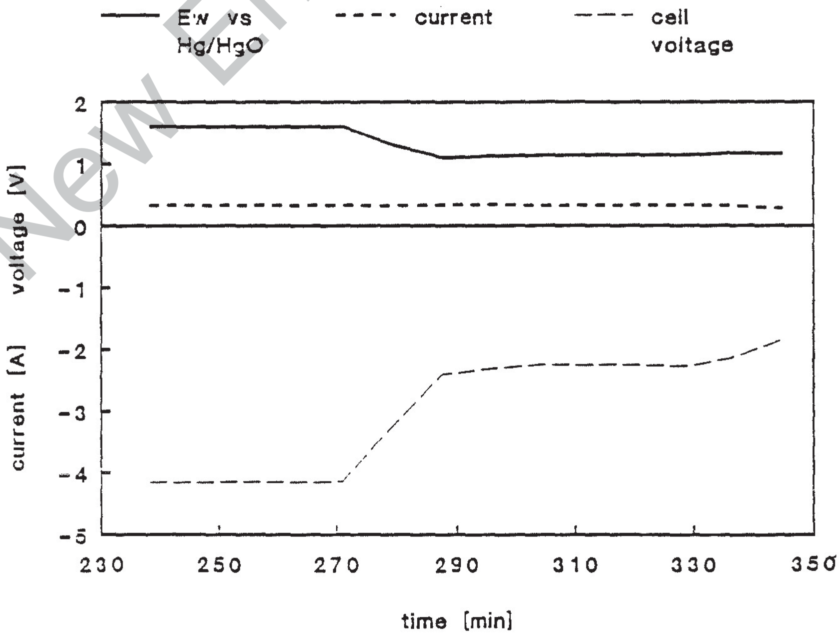


Fig. 2 : Excess heat production observed in D_2O and H_2O cells



The labels on the left are inverted. Voltage runs from 2 to 4 and current runs a fraction of an Amp.

The drop in voltage shown here concurs with the text in the body of their paper.

Fig. 3 : DC measurements while excess heat production was observed with the D_2 cell (results obtained with H_2O cell are similar)

For those reasons, the search was continued mainly in the direction of a hidden energy input e.g. an alternating current.

4. EXCESS HEAT PRODUCTION SIMULATION

Ostensible excess heat production may occur if an alternating current is superposed on the direct current which feeds the cell.
The probability that this will be induced by the AC from the mains is very low since the DC power supply equipments are very well protected against this. An examination of the potentiostats and the pulse generators did not reveal any defects.
An other possibility is that the electrical circuit approaches an oscillation instability.
An experiment was performed where an AC current with varying frequency was superimposed on the DC current. For a frequency of 50 Hz the obtained measurements were quite well in agreement with those observed in the June experiment. This component is not measured in standard electrochemical experiments since the DC measurement equipment integrates over 20 ms.
A computer simulation of the electrical circuit, shown in Fig. 4, composed of the power source, the electrochemical cell and the feedback through the reference electrode (Hg-HgO) was performed in order to detect those components which may contribute to oscillation. This study showed that for the configuration used in the experiment an oscillation could be induced by a cascade of two integrations, one related to the working electrode and one to the feedback circuit.
Based on those results, experiments have been performed on simulated and real cells. In the real cell the palladium was exchanged by platinum.
The following parameters were varied :

- the capacitance (Cref) at the reference input of the potentiostat;
- the resistance of the electrolytic bridge (Rref);
- the resistance of the connection (lead + contact)(Rcon) between the working electrode and the "working" input of the potentiostat.

The results obtained with the real cell are shown in table 2.
With a well functioning cell (good connections and low resistance of the electrolytic bridge) oscillation occurs with a capacity Cref of at least 0.2 μF near 80 Hz.
This frequency lowers to near 50 Hz for a Cref of 0.47 μF.

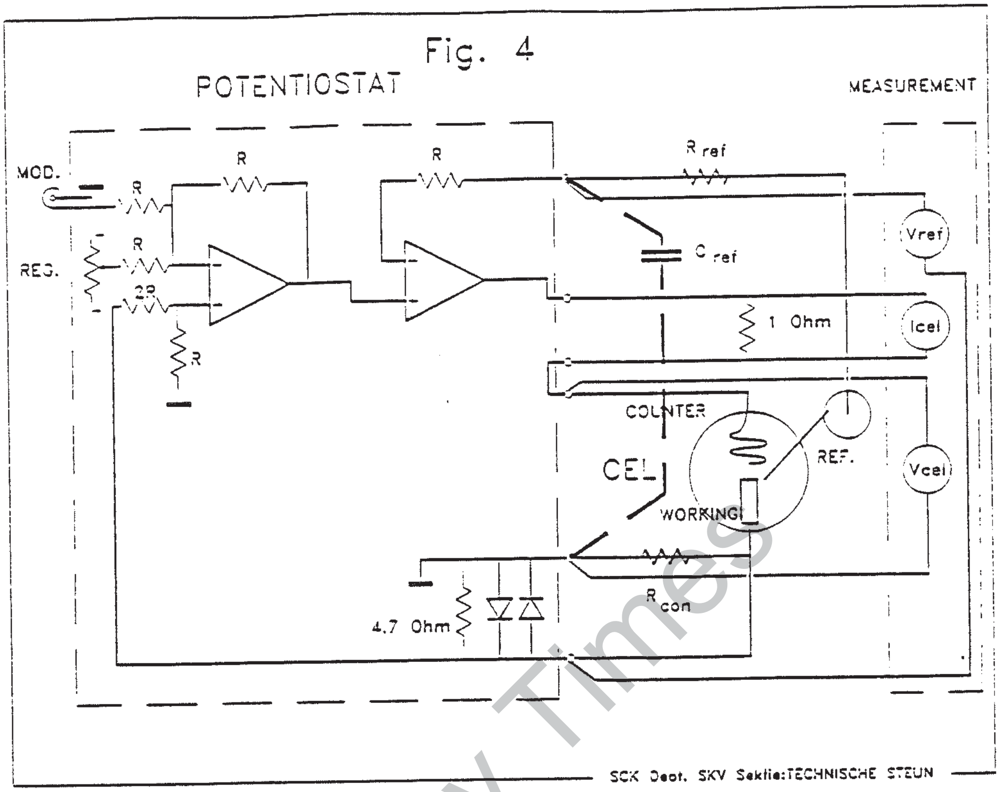


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Fig. 4 : Electrical circuit of the connection of the potentiostat to the working cell (CEL) and the reference electrode (ref.). The extra resistances Rref and Rcon and the capacitance Cref are also shown.

	Single cell				Configuration with 2 cells in parallel
Rref	normal	normal	high	high	high
Rcon (cable to working cell)	normal	increased	normal	increased	increased
Cref (for oscillation)(μF)	0.2	0.1	0.07	0.02	0.01

TABLE 2

Experimental results giving rise to oscillations

In case a bad connection (resistance of a few ohm) occurs between the working electrode and the potentiostat, the required capacitance for oscillation lowers to about 0.1 µF.
If the resistance in the electrolytic bridge increases to a high value (e.g. 2 MOhm) and the rest of the cell is in good working condition, the cell continues to function properly. In this case a capacitance Cref of 0.07 µF is sufficient to induce oscillation (at 32 Hz).
If on top of this also a bad connection occurs as mentioned in a previous case, the required capacitance lowers to 0.02 µF for full power oscillation at 15 Hz and to 0.003 µF for low power oscillation at 58 Hz.

In the actual experimental set-up for the cold fusion experiments the modulation inputs of the potentiostats of the two cells were coupled to the pulse generator. The screening of the connection cable is connected to the sensitive side of the input. This particularity of the modulation input of the potentiostats yields an electrical coupling between the two cell systems. It facilitates pick-up of external signals. In this case only a high resistance in the electrolyte bridge is sufficient to detect a considerable 50 Hz signal on top of the DC potential.
If in this case also a bad connection of the working electrode and a high resistance in the electrolytic bridge occurs, the required capacitance for full power oscillation is lowered to 0.01 µF at 50 Hz.

5. DISCUSSION

Based on the electronic analysis of the systems, experimental evidence is given that a 50 Hz component may become superposed on the DC potential if some "faulty conditions" (perhaps supported by changing electrode capacitances) occur at the same time in the electrical circuit of the cell. This AC component will not be detected by the DC measuring equipment.
The required capacitance, Cref, may be build up partially by the electrical connection lead between the power supply and the cell and a capacitance at the reference electrode.
The high resistance at the reference electrode, Rref, can be caused by insufficient wetting of the electrolytic bridge or the presence of a gas

bubble. The cell continues to function properly, even with an increased resistance in the feedback to the reference electrode.

In case the near instability conditions have been established, the oscillation may be triggered by an 50 Hz perturbation coming from an external source.

The actual energy input due to the 50 Hz AC component depends on the values of Cref, Rref and Rcon.

6. CONCLUSION

Excess energy has been observed during a Fleischmann and Pons type cold fusion experiment.

This phenomenon may be explained on the basis of a hidden input of electrical energy, which is not revealed by the standard DC measurements done in this type of electrochemical experiments.

The hidden electrical energy consists of a low frequency AC power which is superimposed on the applied DC power. It may be generated by the occurrence of an oscillation instability in the electrical circuitry of the cell in case certain faulty conditions build up.

The actual excess heat production phenomenon could be reproduced in simulation experiments.

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REFERENCES

[1] Fleischmann M., Pons S., "Electrochemically induced nuclear fusion of deuterium", J. Electroanal. Chem., 261 (1989) 301-308.

[2] Appleby A.J. and Srinivasan S., DOE Cold Fusion Workshop, Santa Fe, NM, May 25-28, 1989.

[3] Huggins R. and Scherber M., DOE Cold Fusion Workshop, Santa Fe, NM, May 25-28, 1989.

[4] Kainthla R.C., Velev O., Kaba L., Lin G.H., Packham N.J.C., Szklarczyk M., Wass J.C. and Bockris J.O.M., (Texas A&M University).

[5] Kainthla R.C., Szklarczyk M., Kaba L., Lin G.H., Velev O., Packham N.J.C., Waas J.C. and Bockris J.O.M., Eight chemical explanations of the Fleischmann-Pons effect. Department of Chemistry, Texas A&M University.

[6] Interim Report of the Cold Fusion Panel to the Energy Research Advisory Board (US-DOE).

[7] Hydrogen in Metals I and II, Topics in Applied Physics, Volumes 28 and 29, eds. G. Alefeld and J. Völkl, Springer Verlag, 1978.

[8] Dandapani B., Fleischmann M., "Electrolytic separation factors on palladium", J. Electroanal. Chem., 39 (1972) 323-332.

[9] A. Bruggeman, R. Craps, R. Leysen, F. Poortmans, C. Van der Poorten and M. Snykers, "In search of cold fusion during the electrolysis of D_2O. A survey of the experimental work performed at the Belgian nuclear research centre SCK/CEN in the period April - June 1989". SCK/CEN report. July 1989.