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Research Article

Heat Production and RF Detection during Cathodic Polarization of Palladium in 0.1 M LiOD

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Abstract

The study of the Fleischmann–Pons Effect (FPE), i.e. the production of excess power production during electrochemical loading of deuterium in palladium (in the past labeled cold fusion) has had a notably multidisciplinary character, involving solid state and nuclear physics, material science, electrochemistry and other fields. Correlations between the material status and the occurrence of the effect as well as some changes in the electrochemical interface status have been observed. Although during the last two decades, in several calorimetric experiments the effect was observed to be well above the measurement uncertainty, the mechanism producing the excess power is not completely understood. So far, the lack of reproducibility has been responsible for the absence of a clear explanation of the phenomenon based on experimental apparatus designed to enhance the spectrum of information required to define the effect. Recently, In order to improve this aspect of this research, specific work has been carried out to investigate whether the excess power was produced concurrently with the emission of Radio Frequency from the active cathode. Suitable probes and triggering included in the RF experimental system revealed RF signal emission during electrochemical loading of palladium samples, and a correlation between the heat production, in active samples, and RF signal emission was found. The preliminary results highlight the importance of performing studies on the electrodynamic effects that may be involved in the phenomenon, and the importance of the design of appropriate instruments designed to investigate unexplored regions of metal hydrogen systems.
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Keywords: Anomalous heat effect in metal hydride, Electrochemistry, Electromagnetic stimulation, Fleischmann and Pons effect, Metal hydrides and deuterides, Palladium, Palladium alloys, Radiofrequency

1. introduction

The excess power effect was observed using both Pd and Pd90Rh10 cathodes [1]. Some specific features were identified for the active electrodes with both materials [2–4]. The active Pd electrodes had mostly (100) orientation of the poly-crystals, enhanced hydrogen (deuterium) diffusivity, average grain size around 100 μm and a well-defined surface morphology identified by means of the power spectral density function (PSD).

Active Pd90Rh10 cathodes have a high diffusivity and a surface morphology, in terms of PSD [4], very similar to the one belonging to the active Pd samples. The differences are a mixed orientation of the crystals and a lower

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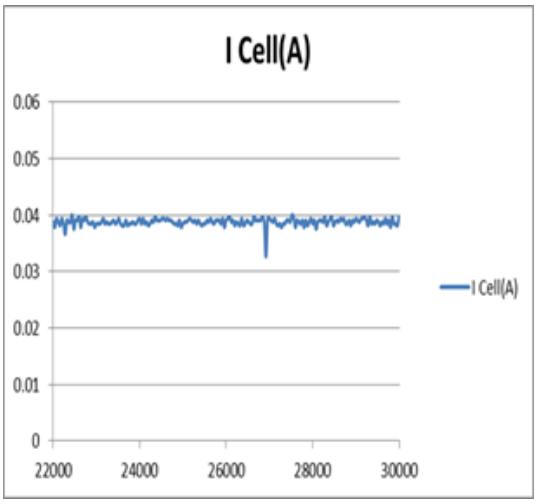

Figure 1. Current ripple during excess power at low data acquisition rate.

grain size, typically some tens' of microns. Doping with contaminants has been performed for both Pd and Pd90Rh10 cathodes in order to achieve the material characteristics belonging to samples producing excess power.

The current ripple observed during galvanostatic loading of deuterium in Pd cathodes led us to consider whether some RF emission from active cathodes occurs during excess power.

Figures 1 and 2 show the current evolution during the excess. Figure 1 shows a ripple at low data acquisition rate (sampling interval 40 s), the ripple in this case was ascribed to an under sampled signal. Figure 2 also shows current and voltage signals acquired at a high data acquisition rate: clear current and voltage oscillations are seen.

2. Experimental Results

This evidence supported the hypothesis that a radio frequency signal was produced during the event; the first set of experiments was performed [5], and signals up to some hundreds of megahertz were detected.

A series of experiments were designed to investigate a higher frequency range up to some tens of gigahertz [6]. During this set of tests a magneto-acoustic trigger was also used, and the RF signal was detected by using a down-converter, located into the electrolyte, and connected to a National Instrument RF spectrometer.

Both the magnetic trigger and the down converter were provided by an anonymous electronics company. During the magnetic stimulus, a clear RF signal was revealed by the RF spectrometer and the electrolyte temperature started to increase by several degrees Celsius in about 20 min. The trigger was disconnected as soon as the temperature increase was observed.

Figure 3 shows the evolution of the cell temperature during the excess heat production. Figure 4a shows the observed RF signal during the event and Fig. 4b shows the typical background RF signal for inactive samples.

The calorimetric approach followed during this experimental series was based on a simple isoperibolic temperature calibration. During this experimental series active and inactive lots of palladium cathodes have been identified and active lots were obtained by using a specific metallurgical procedure.

A further series of experiments were carried out by using a Peltier calorimeter. One of the electrodes, which was from one of the lots that produced active behavior during the experimental series described above, showed clear excess power. The calorimetric behavior is shown in Fig. 5.

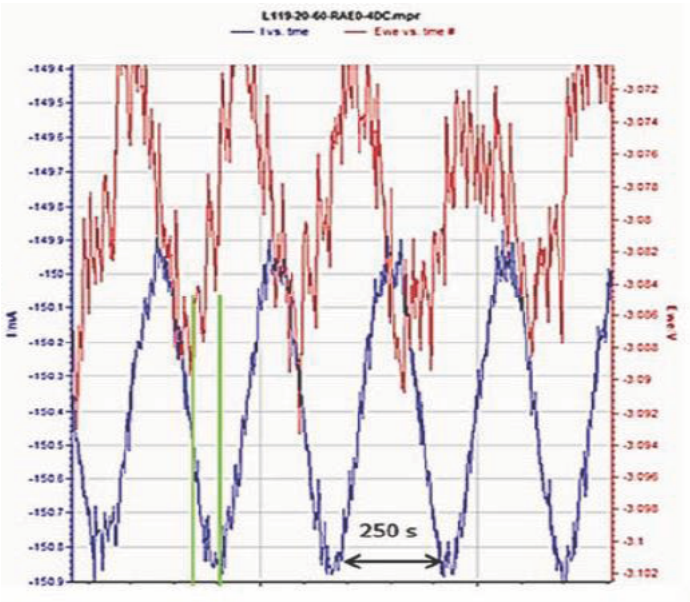

Figure 2. Current ripple at high data acquisition rate.

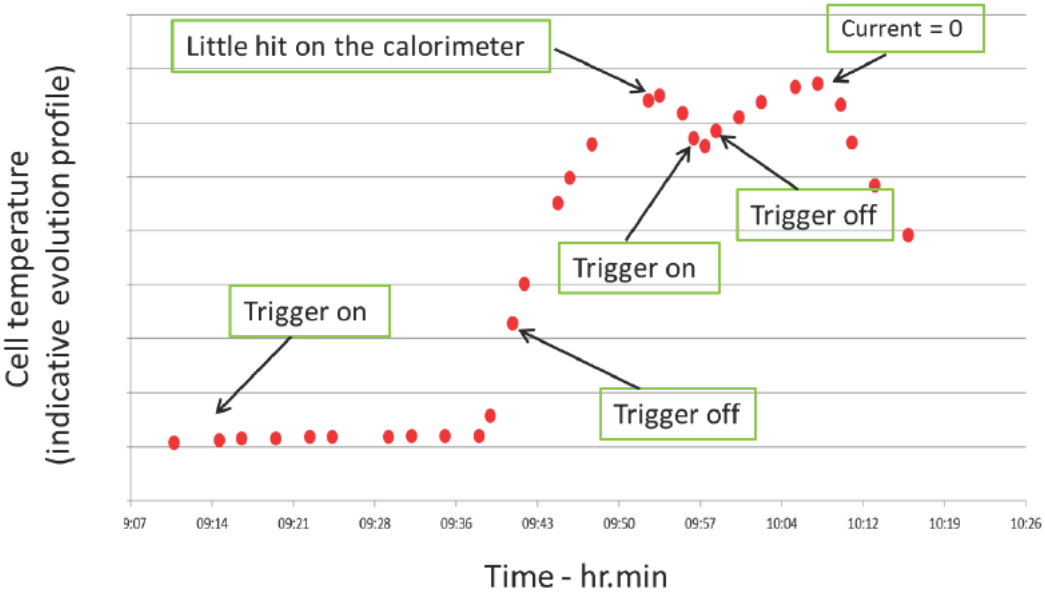

Figure 3. Temperature evolution at the cell wall.

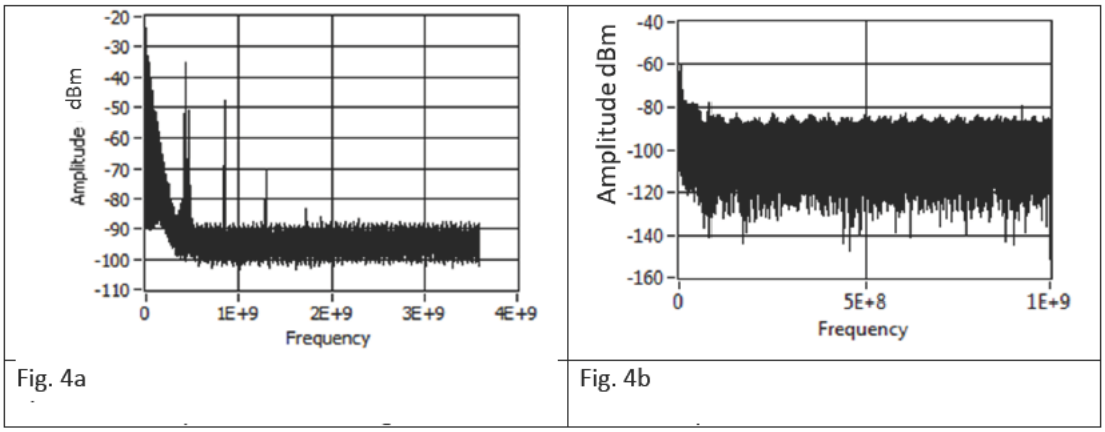

Figure 4. (a) Spectral RF signal for active sample when electrolyte temperature increases. (b) Typical Background RF signal for inactive samples.

The Peltier calorimeter was constructed from a thermally insulated aluminum pipe containing the electrochemical cell. The Peltier cell was located between the bottom of the pipe (not thermally insulated) and an heat sink. The calibration was based on the voltage signal given by the Peltier cell and the following relationship was used to measure the cell output power:

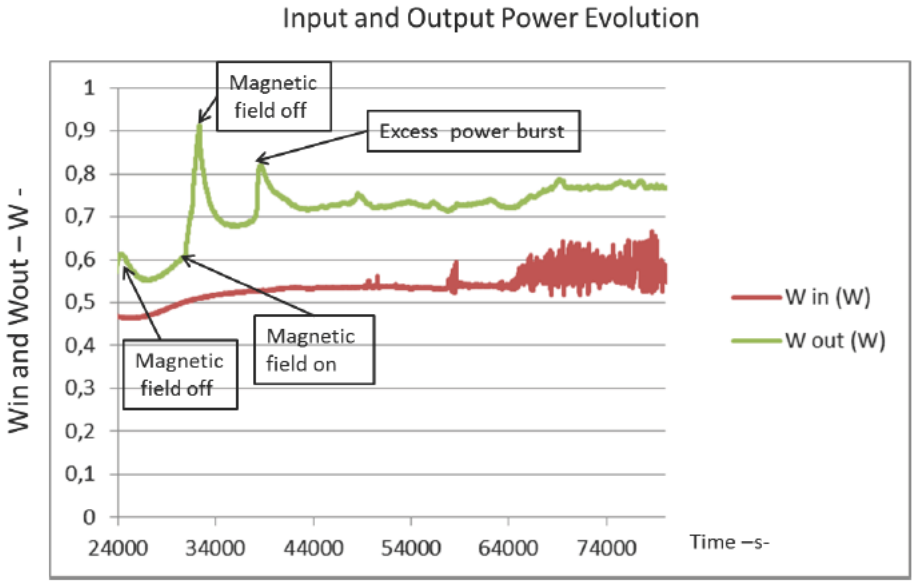

Figure 5. Input and output power evolution during excess power measured by using a Peltier calorimeter. Spikes in input power starting at ∼50,000 s and increasing in amplitude and frequency at ∼65,000 s were due to voltage fluctuation. This effect is not reflecting in the output power because of the calorimeter time constant, which is much larger than the fluctuation frequency.

$$P_{\rm out} = a\,\Delta V + b,$$

where ΔV is the Peltier cell voltage, a and b are coefficients given by the calibration performed with light water (LiOH). The input power in DC current mode was just the scalar product between the current and voltage since the cell used was a closed one.

We may observe in Fig. 5 that after applying the magneto-acoustic trigger the output power ($W_{\rm out}$) exceeded the input power ($W_{\rm in}$). The excess was about 0.1 W when the input power was 0.5 W. The trigger was applied again for several minutes and then removed. After this operation an excess power burst occurred. The plot shows the maximum value of the difference between the output and input power was at least $\sim$0.3 W when the input power was 0.55 W. The burst duration was too short compared to the calorimeter transient time to be measured accurately. Clearly it was much larger than 0.3 W. After this event the excess continued at an average rate of 0.2 W, and after several hours the experiment was switched off and the electrode removed in order to analyze the sample.

Table 1 shows a summary of the experiments performed. We see that only samples belonging to lots 122 and 137 (produced by using the same preparation protocol to have the same metallurgy) were active.

3. Conclusion

RF signal emission has been observed during excess power production. However, such a signal has also been obtained, with the same amplitude observed during excess power, when excess power was not observed. The detection limit of the isoperibolic calorimeter used during RF measurements was of the order of 100 mW. The excess power observed during RF emission was well above this detection limit .

Table 1. Summary of experimental results. Unless otherwise specified, anodes are of stainless steel coated with platinum.

Electrode	Lot	Electrolyte	Current mA	Notes	RF	Heat
Steel	NA	H_2O	53	NA	No	No
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L124(10–50)	Pd90/Rh10 (Al)	H_2O			No	No
L127(20–130)	H.M. + Pt	H_2O			No	No
L128(105–145)	H.M. + Pt	H_2O			No	No
L128(145–185)	H.M. + Pt	H_2O			No	No
L58(165–200)	Johnson Matthey MM29560	H_2O			No	No
L93(110–150)	Alfa Aesar 307622	H_2O			No	No
L95(50–90)	Alfa Aesar 307622	H_2O			No	No
L121(90–130)	Pd90/Rh10	H_2O			No	No
L125(170–206)	H.M. 2108539 + Pt	H_2O			No	No
L122(120–160)	Pd+Doping	H_2O	24		Yes	Yes
L122(120–160)	Pd+Doping	H_2O	53		Yes	Yes
L122(160–200)	Pd+Doping	H_2O	53		Yes	Yes
L122(160–200)	Pd+Doping	H_2O	53	Charge 107 mA	Yes	Yes
L122(236–272)	Pd+Doping	H_2O	53		No	No
L122(308–366)	Pd+Doping	H_2O	53		Yes	No
L137(100–140)	Pd+Doping	H_2O	53		No	No
137(180–216)	Pd+Doping	H_2O	107	Anode Pt	No	No
L137(60–100)	Pd+Doping	H_2O	107	Anode Pt	Yes	No
L137(60–100)	Pd+Doping	H_2O	107	Anode Pt	Yes	Yes
L137(300–340)RAEM	Pd+Doping	H_2O	107	Anode Pt	Not measured	YES
L137(380–420)RAEM	Pd+Doping	H_2O	107	Anode Pt	Yes	No

This indicates that RF emission is not the effect of the excess, but rather, perhaps the cause. Electrochemical instability is observed coinciding with the onset of the effect. Active electrodes have been obtained by doping a rough palladium sample that was originally inactive. Such a result should be considered as preliminary, since a more significant statistical basis of data is required. A further effort in this direction could improve the control of the effect.

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