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Mechanism of thermal runaway as a cause of Fleischmann-Pons effect

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Abstract

Starting from papers by Fleischmann and Pons, many investigators have found the excess power effect during a heavy water electrolysis. They connected this effect with the deuterons "cold fusion". A significantly larger number of investigators did not have found this effect, so they do not agree with the proposed explanation and consider the results of Fleischmann and Pons being a mistake or an instrumental artifact. In this paper experimentally proved that the Fleischmann-Pons effect (of burst type) is caused by an exothermic reaction of a recombination of the atomic deuterium accumulated in electrodes during electrolysis of the electrolyte. This reaction is similar to the reaction of thermal runaway in electrochemical batteries with aqueous electrolyte. Thus experimentally proved that the Fleischmann-Pons effect is not associated with cold fusion of deuterium nuclei. While the Fleischmann-Pons effect (of the weak type) is due to a partial recombination of the deuterium and the oxygen, i.e. in this case the excess power is apparent or imaginary. It is shown that the established mechanism of Fleischmann-Pons effect explains all the currently known experimental facts. The recommendations are given allowing a reproduction of this effect without a failure.

Keywords: thermal runaway; Fleischmann-Pons; deuterium accumulation; deuteride; hydride; battery

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1. Introduction

In 1989, M. Fleischmann and S. Pons made a sensational statement [1]. According to their investigations in a case of a heavy water electrolysis with a palladium cathode, the emergence is possible of the excess power effect; i.e. a cell starts releasing more energy than it obtains at the moment from a power source. Due to an absence of evident exothermic (chemical or electrochemical) processes in the cell as well as due to the great importance of the excess power effect, Fleischmann and Pons suggested that an origin of the excess power is a reaction of deuterons fusion. This hypothesis was called by Fleischmann and Pons “cold fusion” or “low energy nuclear reaction” (LENR). Hereinafter many investigators tried repeating the results by Fleischmann and Pons but most part of them did not find this effect [2-4]. This is why, it was then the practice to consider that the results obtained by Fleischmann and Pons are due to some experimental mistakes or an instrumental artifact [5,6].

Nevertheless recently, the excess power effect during a heavy water electrolysis was confirmed by investigators from a number of well-known scientific centers of the world: Energetic Technologies (ET-Omer, Israel), SRI Int. (Menlo Park, CA), ENEA (Frascati, Italy), etc [7-11]. This is why if to analyze all the investigations on the Fleischmann-Pons effect (F-P), one can come to the following conclusions. Firstly, if the effect of the excess power release exists, it is barely occasional and extremely unlikely. Secondly, until now, any well-founded experimental proves do not exist that this effect is due to the reaction of the deuterons fusion, i.e. the “cold fusion”. This fact is recognized even by the most passionate proponents of the F-P effect [11].

If however still presume a possibility of the “cold fusion”, a lot of insurmountable theoretic problems arise connected with explanation of by-effects. Indeed, if a fusion of two nuclei takes place (it does not matter on which mechanism – “cold fusion” or “hot fusion”), a great energy is released, which can be realized either via γ -radiation and/or via big kinetic energy of a products of the reaction. Nevertheless, in the F-P effect process, no high-energy irradiations were found, at least ones sufficient for an explanation of the excess power effect. Besides, until now no reliable theory was suggested,

which could explain, how the releasing of two nuclear fusion energy could be transferred to a metal crystal lattice without a high-energy external radiation [11].

This theoretic problem would not come to existence at all, if to presuppose that the F-P effect is caused by a course of a powerful exothermic (chemical or electrochemical) reaction. But at present also no mechanism has been proposed for an explanation of the F-P effect based on any chemical or electrochemical reactions.

Judging on many external signs the F-P effect is similar to the effect of the thermal runaway in electrochemical batteries with an aqueous electrolyte [12-22].

Firstly, both of these effects are occasional and extremely unlikely [13-18]. Secondly, as a result of both F-P effect and thermal runaway, the energy releases much larger than an electrochemical cell obtains from a power source at the same time [13,15]. Thirdly, both these effects arise at electrolysis of an aqueous electrolyte.

According to the standard point of view [12] the thermal runaway in batteries goes on with the following scenario. In a case of a battery's long recharging under a constant voltage (or at its floating service), heating-up takes place, which results in decreasing of a battery's internal resistance and in an increase of the recharging current, which in its turn magnifies heating up, etc. So the thermal runaway is a result of a positive feedback between a current and a temperature of batteries during their charging under a constant voltage.

So the classical mechanism of the thermal runaway supposes that a cause of the thermal runaway is an acceleration of electrochemical reactions of batteries charging and a decomposition of an electrolyte (as the thermal runaway takes place in a case of a long-lasting recharge of batteries) on expense of batteries heating and decrease of their internal resistance.

In our previous papers [15,16], there was shown that a lot of experimental data exists contradicting to the generally accepted mechanism of the thermal runaway [12]. For example, according to the classical mechanism [12], the thermal runaway occurs on expense of an energy obtained by a battery from a recharger. But the direct experimental studies show that as a result of a

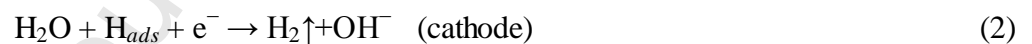
thermal runaway, a battery releases energy approximately 140 times more than it obtains from the recharger [15]. This fact can be explained only from an assumption that the thermal runaway is accompanied by a powerful exothermic reaction. Moreover, this reaction must be a basis of the mechanism of the thermal runaway.

On the basis of all the available experimental data in the papers [15,19], there was proposed the new thermal runaway mechanism. In these papers, there was shown that there are two processes of accumulation that step-by-step bring the nickel-cadmium batteries to the thermal runaway. Firstly, this is a process of a hydrogen accumulation inside of battery electrodes during their operation. Secondly, this is a process of dendrites accumulation on the cadmium electrodes of the nickel-cadmium batteries. In the papers [14,16] by a thermal decomposition of electrodes, there was shown that in a course of a long operation (more than five years) of the batteries KSX-25 (with sintered electrodes by capacity 25 Ah), in their electrodes, up to 800 liters hydrogen are accumulated.

In the paper [19] there was shown by experiments that an exothermic reaction of the thermal runaway in nickel-cadmium batteries is the reaction of a recombination of accumulated atomic hydrogen

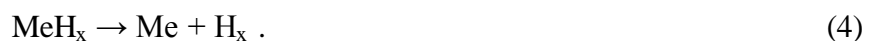


which runs in line of electrochemical mechanism



Hydrogen is present inside of battery electrodes in a form of metal-hydrides, i.e. in atomic form [23]. The reaction (1) is a powerful exothermic reaction with heat emission in amount of 436 kJ/mole (hydrogen) [24]. This heat emission is more intensive than in a case of hydrogen combustion reaction in oxygen, which makes 285.8 kJ/mole (hydrogen) [25,26].

A rate-limiting step for reaction (1) on both a cathode and an anode is found to be a step of a metal-hydrides disintegration [19]



In a place of a dendrite development, a current density of a battery charge will be much higher than at other places of an electrode as the distance to a counter electrode is less. Hence, this place will be heated up much more intensively than other electrode parts. A reaction rate of hydrides decomposition (4) increases with a temperature growth. As the reaction (4) is a rate-limiting step for thermal runaway reactions (2,3), in proportion, an intensity of those powerful exothermic reactions will grow and consequently the electrode will be heated up even more, and so on (Figs. 1,2) [15].

In the experiments (Fig. 1), KSX-25 batteries were charged at a constant voltage of 1.87 V. A thermal runaway can occur in batteries only in a case of their long-time recharge [12]. In our previous paper [19], it was shown that to a moment of a thermal runaway initiation in the battery KSX-25, the recharging current was very small, about 0.38 A (Fig. 1). Dominantly, it was consumed on electrolyte decomposition and in a much lower percentage on the battery charge (as it was mode of recharging). The reason of the recharging current value growth (Fig. 1) is connected with electrochemical reactions occurrence (2,3) in accordance with the mechanism of thermal runaway described above [19].

The exothermic reactions (2,3) requires new hydrides for its continuation. In a spot of the reaction initiation (2,3), they will be exhausted; and reaction will shift radially from this point burning a separator in a form of regular circles, which indeed is observed in the experiments (Fig. 2).

An interruption of the electrochemical reactions of the thermal runaway (2, 3), is connected with electrolyte evaporation between electrodes (as a result of high temperature). This gas lock will gradually physically interrupt electrochemical reactions of the thermal runaway (2, 3) and decrease the recharge current (Fig. 1).

In the papers [15,19] it was shown that the standard potential difference between the cathode and anode for thermal runaway reactions (2,3) is approximately 0.5 V. Therefore, when electrochemical reactions of thermal runaway (2,3) occur, the voltage at the battery terminals drops to 0.5-0.6 V, since in this case the contribution of other electrochemical reactions is extremely insignificant (Fig. 1). After the end of thermal runaway, the voltage at the battery terminals returns to its original value, if there is

no short circuit of the electrodes. It should be noted that in nickel-cadmium batteries, thermal runaway can be represented by several energy bursts (Fig. 1.2(b)) and [14,19].

In the paper [19] it was shown that this mechanism of the thermal runaway explains all currently known experimental facts.

In this paper we explore the F-P effect based on this thermal runaway mechanism.

2. Experimental

Many investigators studying the F-P effect [10,11,13] point out that the excess energy can be of two types.

(Type A). The excess energy releases instantly in a form of a burst. In this case, the power released by a cell is superior several times to the power received at the moment by the cell from a power source. This type of the excess energy emerges very seldom and it is observed only after several weeks or months of electrolysis. This type of the excess energy can not be explained by any experimental mistakes.

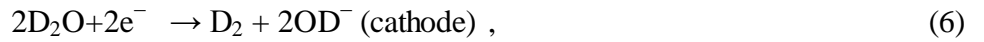
(Type B). In this case, also the power released by a cell is superior to the power received at the moment by the cell from a power source but rather insignificantly. The excess energy of this type emerges on 2nd or 4th day after the electrolysis initiation and can be continued during many days. In our investigations, this type of the excess power emerged approximately in 7% of cases. Usually, this type of the excess power is explained by errors of calibrating or other experimental errors [2-6].

The main purpose of this experiments group is repeating of the classical Fleischmann-Pons experiments [1,13] and evaluating of emergence probability of the excess power effect. In connection with this, the used by us cells for heavy water electrolysis exactly reproduced the cells used by Fleischmann and Pons in their experiments [13]. Measurements were carried out in Dewar cells maintained in a large constant temperature water bath (30°C). The palladium cathodes were welded to 0.05 cm diameter palladium lead wires and these leads (as well as those to the platinum anodes) were covered with glass shields in order to avoid any possible recombination of deuterium and oxygen in

the gas head spaces of the cells. Temperature measurements were made with specially calibrated thermistors (Thermometrics Ultrastable Thermoprobes, $\sim 10 \text{ k}\Omega$, $\pm 0.02\%$ stability per year). Besides, at conducting of the experiments and results obtaining, in whole we used the F-P methods described by them in detail in their papers [1,13]. In particular, a cell input power was found on the formula

$$P_{in}=(U-UI_m)I, \quad (5)$$

where U , I are cell voltage and current, $UI_m=1.54\text{V}$ is thermoneutral voltage for electrochemical reactions of the electrolyte decomposition



At the voltage UI_m , the supplied energy is spent only for the electrolyte decomposition, while as for Joule heat, it does not release in a cell. The power output was found according to the method by F-P described in detail in the paper [13].

For enlarging of experimental results statistics, we used two baths of water at 30°C . Each bath contained five Dewar's cells. This way, we worked with ten cells at the same time. As a cathode, there was used a palladium rod (99.9%) 0.4 cm in diameter and 1.25 cm in length. The electrolyte was a solution of 0.1M LiOD in heavy water (99.7% isotopic purity). The electrolysis current density was 64 mA cm^{-2} . As an anode, there was used a platinum wire (99.9%) 0.1 cm in diameter. The helical platinum anode was tightly wound with a close spacing on cage of glass rods surrounding the cathode. These parameters of the cell were absolutely equivalent to the parameters used by Fleischmann and Pons in their experiments, in which they obtained the excess power (of type A) [13].

The constant current was supported with aid of a potentiostat (HiTek Instruments Model DT2101) in the galvanostatic mode.

3. Results and discussion

3.1 Fleischmann-Pons effects

The electrolyte decomposition was performed in average during 90 days. In total, ten experiments cycles were conducted, apart from necessary cells calibrating operations. So the total number of experiments makes $10 \times 10 = 100$. As a result of the experiments conducted, only once, we observed the F-P effect (of type A) (Fig. 3 and Table 1, experiment #71) and the F-P effect (of type B) seven times.

All the results of the conducted experiments are represented in the Table 1. In this Table we show parameters only for one of the seven observed by us F-P effects (of type B, experiment #65) as the parameters for the rest six effects (of type B) were similar.

In many papers on investigation of the F-P effect (of type B) there was proved that with growth of the current density, the excess power grows, too [10,11,13]. This is why, we conducted a separate group of 10 experiments under the current density 256 mA cm^{-2} , while using $0.2 \times 1.25 \text{ cm}$ Pd rod in the same electrolyte. As a result of the conducted experiments, we observed the F-P effect (of type B) only once (Fig. 4 and Table 1, experiment #107).

According to the conducted experimental investigations, the emergence frequency of the F-P effect (of type A) was 1%, while of the F-P effect (of type B) 7%. In the experiments described in the paper [16], the emergence frequency of the thermal runaway in the KSX-25 batteries was equal to 0.125% at charge voltage 1.87V and temperature 25°C . In real operation conditions, the emergence frequency of the thermal runaway in these batteries is much lower as they are operated in floating mode at the voltage not higher than 1.45V.

One should draw attention that at the moment of the thermal runaway emergence, the current density of the KSX-25 battery charging was 40 times lower [19] than at the moment of the F-P effect emergence (of type A) (Fig. 3). Although as it is known, a probability of both the thermal runaway and the F-P effect grows with increase of the current density [11,16]. So based on the conducted experiments, at least, one can affirm that both effects are extremely unlikely. It is necessary to

highlight also that not in every group of such experiments, we succeeded in obtaining both the thermal runaway and the F-P effect (of type A), i.e. both these effects are sporadic and depend on many factors.

3.2 Fleischmann-Pons effect (of type A)

Based on the conducted experimental investigations in the papers [14-19], we presupposed that the cause of the F-P effect (of type A) is the exothermic reactions of the thermal runaway (2,3), which – in the case of alkaline electrolyte based on heavy water – should be written in the following form:



The overall reaction:



A rate-limiting step for this reaction on both a cathode and an anode is the step of metal-deuterides disintegration [19].



In connection with this in course of the F-P effect (Fig. 3), into a measuring container, we collected 3106 ml gas. The gas composition analysis showed that 99.5% of it consisted of deuterium. So the volume of the released deuterium (not compensated by oxygen) will be 3059ml. With due account to the fact that a heat emission at the recombination reaction of atomic deuterium is equal to 442.4 kJ/mole(deuterium) [27], we obtain that deuterium evolution from electrodes must be accompanied with release of 60609 J of heat. In limits of error 1.3%, this value coincides with calorimetric measuring of the heat generated from the cell during the F-P effect (of type A) (Table 1, experiment #71).

Thus, the conducted experimental investigations strongly prove that the cause of the F-P effect (of type A) is the exothermic reaction of the thermal runaway (10).

3.3 Electrodes of F-P cell as deuterium storage units

Now let us estimate a capacity of F-P cell electrodes as deuterium storage units. With this purpose we conducted a separate set of experiments using electrodes from the experiment #71 (with use of which the F-P effect (type A) was observed) and electrodes from the experiments #54 and #107 (upon use of which the F-P effect (type A) was absent).

In cells with these electrodes, the decomposition was performed of the same electrolyte by the current density 30 mA cm^{-2} during 100 days. Then the electrodes were subjected to thermal decomposition in a hermetic thermal chamber at the temperature 800°C . The released (as a result of thermal decomposition of the electrodes) gas passed through a coil-in-box cooler and was collected into a measuring container. In detail, this installation and experimental method were described in the paper [14]. The experiment results are shown in the Table 2.

From the Table 2 analysis, two conclusions can be made. Firstly, the electrodes #71(Pd and Pt) are able accumulating deuterium and even much more than it is needed for the F-P effect (type A) (Section 3.2), on the basis of the thermal runaway mechanism (8,9). Secondly, at electrodes from different commercial consignments, a gravimetric capacity of deuterium accumulating differs very much.

The electrodes #71(Pd and Pt) were from the oldest consignment. They were received by us as long ago as in nineties and previously many times they took part in similar experiments. The electrodes #54 (Pd and Pt) and #107 (Pd and Pt) were from the last consignments received by us during the last five years especially for these experiments conducting.

In paper [28], it was experimentally found (using Scanning Electron Microscopy and Transmission Electron Microscopy) that in electrodes, where the F-P effect (of type A) was observed, a density of micro-defects and dislocations was much higher than in electrodes, on which this effect had never emerged. In the papers [10,29], the analysis was conducted of palladium electrodes from different consignments for many years. It was noticed that from mechanical point of view, the electrodes used in historically successful F-P experiments were less solid than the electrodes supplied

by the same company during last years. Although from these experiments, authors made no conclusions.

We checked the relationship established in paper [28] between the number of micro-defects and dislocations in the electrodes and the amount of deuterium accumulated in the electrodes.

The Scanning Electron Microscopy of the samples (Table 2) showed that on the surfaces of the electrodes #71 (Pd and Pt), there were a lot of lots of splits and other micro-defects, meanwhile, for example, on the surfaces of the electrodes #54 (Pd and Pt), any micro-defects were very rare and notably that on many parts they were absent at all.

The Transmission Electron Microscopy of the samples (Table 2) showed that, for example, in the palladium cathode #71, the density of dislocations was eight times higher than in the palladium cathode #54. As for the corresponding platinum anodes, this ratio is a little bit less (approximately 7.2).

Nevertheless, it is evident that on their structure, those electrodes that contain a lot of micro-defects and dislocations are closer to metal-ceramic electrodes used in alkaline batteries.

In the papers [14,30,31], it was proved by experiments that in order that in metal-ceramic electrodes of nickel-cadmium batteries, there would be accumulated an enormously big volume of hydrogen, two factors are important: the presence of a lot of micro-defects in electrodes (pores, dislocations, etc.) and a very long duration of an electrolyte decomposition and hydrogen accumulation processes. In the paper [16], it was shown that battery KSX-25 electrodes collect the hydrogen maximal volume only within five years of operation. In a course of a battery operation, the hydrogen is accumulated in its electrodes only during its charging, when an electrolyte decomposition takes place. If to charge a battery constantly, the maximal volume of hydrogen accumulation will be reached by its electrodes for a less period of time than five years. Separate experimental investigations showed that at charging rate 1.5A, the battery KSX-25 electrodes can reach the maximal volume of hydrogen accumulation approximately during one year.

In the paper [30], it was shown that a gravimetric capacity of metal-ceramic nickel matrices of battery KSX-25 oxide-nickel electrodes is equal to 20.1 wt%. The capacity obtained exceeds

previously obtained results for any reversible metal hydrides (obtained by traditional methods), including those for magnesium hydride or complex hydrides by 3 times [23,32].

In a separate set of experiments, we studied impact of electrodes porosity on gravimetric capacity of hydrogen accumulation. With this purpose, several oxide-nickel electrodes of the battery KSX-25 were replaced by electrodes made of compact nickel of the same area and weight. Then after a year-long battery charging by the current 1.5 A, by the way of the electrodes thermal decomposition (according to the method described in the paper [14]), their gravimetric capacity was found. The gravimetric capacity of the electrodes of compact nickel was found to be equal to 0.9 wt%, meanwhile for metal-ceramic nickel matrices of oxide-nickel electrodes, it was equal, as usual, to 20.1 wt%. So the porous structure of the battery metal-ceramic electrodes is the critical factor for the accumulation of a lot of hydrogen in electrodes.

Of course, the phenomenon of accumulation of a lot of hydrogen in metal-ceramic matrices under condition of a long electrolysis requires further both experimental and theoretical investigations. Nevertheless the fact of this phenomenon itself was established by us reliably for as long as 25 years of investigations [30,31].

Micro-defects and dislocations are present in electrodes #71 (Pd and Pt) in large numbers and are almost absent in electrodes #54 (Pd and Pt) and #107 (Pd and Pt). Hence, the main cause of the fact that the F-P effect is observed at use of one consignment of cathodes and is not at use of other similar consignment [11] consists in the electrodes structure. Our many-years experience of such investigations shows that the palladium (in average) from older consignments (20-25 years ago) contained more micro-defects and dislocations than the palladium from modern consignments. It means that the modern palladium is of higher quality, which reduces intensely a probability of the F-P effect emergence. Besides, we noticed that in a course of electrodes long operation, the micro-defects and dislocations in them are accumulated. In the papers [33,34], it is also highlighted that in a course of PdD operation, micro-splits are formed in electrodes.

In the experiment #71 (Table 2), the electrolyte was decomposed by the current 47 mA during 100 days. Consequently within this time, approximately 47 liters deuterium and 23.5 liters oxygen released. This deuterium volume exceeds 12 times the deuterium volume accumulated in the electrodes (Table 2). The similar picture was observed also in the experiment represented by Fig. 3. Prior to the F-P effect emergence, the electrolyte was decomposed by the current 100 mA during 912.4 h. So within this time, about 38 liters deuterium and 19 liter oxygen released. This deuterium volume exceeds 12.4 times the deuterium volume released as a result of the F-P effect (type A) (Section 3.2). Thus, during electrolyte electrolysis, in electrodes, a necessary deuterium volume could be accumulated. Notably that due to the compact cell design, deuterium released on a cathode could get on both cathode and anode and be accumulated in them. In the investigated cell as a separator between the cathode and anode, a pack was used of glass rods by 1 mm in diameter.

We would like also to attract attention to the fact that both hydrogen and deuterium feature with a very high diffusion permeability. For example, at the temperature 20°C, the diffusivity of hydrogen/deuterium in nickel is approximately 10^{10} times higher than diffusivity of nitrogen or oxygen [23,35]. This feature explain, why upon dissociation of an electrolyte into hydrogen/deuterium and oxygen in a cell, only hydrogen/deuterium accumulates in the electrodes, while oxygen escapes to the atmosphere.

3.4 Deposits on the cathode surface

Let us consider the impact of deposits on cathode surface on the emergence probability of the F-P effect. In the papers [15-19], while investigating thermal runaway in alkaline batteries, we proved that what step-by-step brings a battery to the thermal runaway are the two following processes of accumulation: Firstly, this is the process of hydrogen accumulating in electrodes. Secondly, this is the process of deposits accumulating in a form of dendrites on a negative electrode.

Also the dendrites on a negative electrode promote boosting probability of thermal runaway initiation due to two reasons. Firstly, a dendrite grown up through a separator reduces dramatically the

distance between electrodes, and consequently in this spot, a current density would be much higher than in average on the electrode [19]. This is why this place will be heated up faster than other parts of the same electrode. This promotes rate increasing of hydrides decomposition (4) and consequently in this spot a power grows of exothermic reactions of the thermal runaway (2,3), for which the reaction (4) is a limiting stage, i.e. in this spot of an electrode a thermal runaway starts [19] (Fig. 2).

Secondly, the deposits are represented by deeply destructed crystal structures. However any imperfections of metal crystalline structure are traps for hydrogen, as they decrease the energy of hydrogen atom as compared to their location in normal interstice. Besides they are the centers of hydrogen absorption, and also contribute to hydrogen penetration into the metal depth. That is, in a spot of a deposit formation, the activation energy is much lower for processes of hydrogen/deuterium sorption/desorption, and therefore less energy is required for hydrides/deuterides decomposition. As the reaction of deuterides decomposition (11) is a limiting stage for the exothermic reactions of the thermal runaway (8,9), in a deposit location, those reactions power will be higher than in neighboring parts of the electrodes, where the deposits are absent. In its turn, the exothermic reactions power increase will lead to even more intensive heat-up of this spot, which would result in even greater rate of dendrites decomposition (11), and so on. It means that there exists a positive inverse relationship between the deuterides decomposition reaction (11) and the thermal runaway exothermic reactions (8,9) or, which is the same, between an electrode temperature and energy generated. About this positive inverse relationship, many researchers wrote, who investigated either F-P effect [11] or the thermal runaway effect in batteries [15,16].

Thus, spots of deposits formation on a Pd cathode are those places, where the thermal runaway self-accelerating process can be initiated most easily (8-11). After deuterium burning out in the spot of the deposit formation and in cathode places closest to the deposit, the thermal runaway stops. This way in the spot of a deposit formation, a burst emerges with generation of a great energy and release of deuterium. If deposits on a Pd cathode are located close enough to each other, a thermal runaway initiation in a location of one deposit will heat-up the electrode around the deposit and thereby it will

promote an emergence of self-accelerating mechanism of thermal runaway (8-11) in neighboring deposits. Consequently, deposits density increase on a cathode results in probability growth of the F-P effect. This fact was highlighted by many investigators of the F-P effect [10,11,36]. In the papers [10,36], intentionally, the authors added salts of palladium and nickel into electrolyte for deposits density growth; and they pointed out that this results in a great probability increase of the F-P effect emergence.

Thus, two processes of accumulation bring a cell step-by-step to the F-P effect. Firstly, this is the process of deuterium accumulating in electrodes. Secondly, this is the process of deposits accumulating on a cathode surface.

The fact that a deposits formation on a cathode surface is an important factor for an initiation of the F-P effect, was noticed by many investigators of this effect [11]. In their papers, this process was called by them forming a nuclear-active-environment (NAE). Although as for the role of NAE, they were not able explaining this. From above-mentioned investigations, it is clearly seen that the deposits on a cathode surface form the active environment as they are the necessary condition for the F-P effect emergence. But the deposits are an active environment not for nuclear processes but instead for the thermal runaway process (8,9).

So the F-P effect (type A) represents a set of bursts because of the thermal runaway process (8,9) in different places of a cathode and in different moments of time. Namely this is how changing looks like of excess power generated by a cell in a process of the F-P effect (type A) Fig. 3(a). The concept that the F-P effect represents a number of energetic bursts was noticed by many investigators of this process [11,36-38]. The same energetic bursts on a cathode were also photographed in the papers [36,38].

We would also highlight that changing of different parameters in the form of a set of bursts is typical also for thermal runaway in different batteries with aqueous electrolyte, where there are deposits on the cathodes but powerful dendrites are not formed. For example, this is typical for lead-acid batteries. In batteries, where powerful dendrites are formed (for example, such are nickel-

calcium batteries), the thermal runaway is represented by one to three and very seldom to five energetic bursts (Figs. 1,2) and [14,19].

3.5 Cell voltage during the F-R effect (of type A)

In conclusion of this paper part, let us give consideration to a cell voltage change during the F-P effect (type A), Fig. 3(b). From the Fig. 3(b) and Table 1 it is seen that in the case of the F-P effect initiation, a cell average voltage drops approximately by 0.47 V. Such cell voltage drop was established also in the similar F-P's experiment [13]. This is connected with the fact that the electrochemical reactions (8,9) run at much lower voltage (about 0.5-0.6V (Fig. 1) and [19] than the electrochemical reactions (6,7). But at the same time, the F-P effect is accompanied with an intensive deuterium evolution, which blocks electrodes and increases to a large extent an internal resistance of a cell. The simultaneous action of these two processes influencing cell voltage value in opposite directions results in a compromise voltage (Fig. 3(b)). The cell compromise voltage will depend on constructive features of a cell, a state and a form of electrodes themselves, a distance between the electrodes, a status of separator between the electrodes (whether it is used or not), etc., because all these factors are determinative for a size of a gas film on the electrodes and a size of a gas lock between the electrodes. In a case of an electrodes arrangement with a very small distance between the electrodes (as F-P recommends [13]) and separators use (as in F-P's experiments, in the form of a pack of glass rods), the gas locks formation between electrodes is inevitable.

When a cell temperature is high enough (especially close to the boiling point), additionally, a powerful steam formation takes place. In this case always, the cell voltage during the F-P effect is much higher than the voltage before the F-P effect [39] start. Namely the impact of all the above specified factors on a cell compromise voltage can be an explanation of the observed considerable dispersion of the measured voltage values for various cells at various authors [10,11,13,39]. Upon that, a cell voltage during the F-P effect can be either lower [13] or higher [10,39] than a voltage before the effect initiation.

In alkaline batteries, as a rule, the thermal runaway takes place inside of one of several couples of electrodes. Thereat only between these electrodes (and as a rule, only in a location of the thermal runaway [15,16,19]), a gas lock is formed and a resistance grows. Between other electrodes, the resistance becomes a little bit lower because of the electrolyte temperature growth. This is why, in general, during the thermal runaway, a battery resistance changes insignificantly. Hence, a voltage on battery terminals is set up according to a voltage of the electrochemical reactions (2,3) running (approximately 0.5-0.6V) (Fig. 1) and [19].

3.6 Fleischmann-Pons effect (of type B)

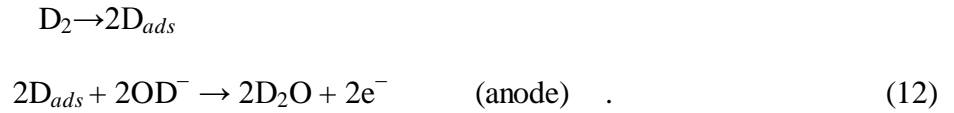
While investigating the F-P effect (of the type B) (Figure 4(a)), also we collected gas releasing from a cell into a metering container.

The gas analysis showed that a ratio between a deuterium concentration and a oxygen concentration makes 2:1, i.e. in a cell only decomposition runs of the heavy water onto deuterium and oxygen. Notwithstanding, if to analyze a gas content released during a small time interval, for example during 0.2 hour, the deuterium concentration can differ from the ratio 2:1 by a value up to 3%. This is indicative of the fact that in the F-P effect process (of the type B), there are parts, on which deuterium release gathers pace because of reactions (8,9) (in the Figure 4(a) to such parts, bursts correspond). And also there are parts, where deuterium is absorbed by electrodes more intensively so that deuterides are formed (in the Figure 4(a) to such parts, dips correspond). In whole over the entire period of the F-P effect (Figure 4(a)), the ratio between the deuterium concentration and the oxygen concentration makes 2:1.

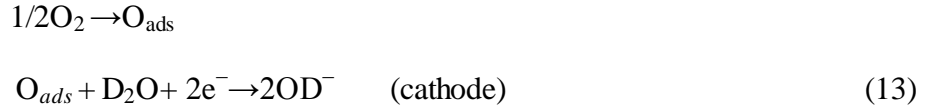
In our experiments, a current efficiency for the reactions of electrolyte decomposition (6,7) was higher than 99%. The same high values of the current efficiency were also obtained in the papers [3,13]. Though after beginning of the F-P effect (of the type B), the current efficiency of electrolyte decomposition started slowly decrease down to the value 81.5% (Figure 4(b)). Such value decrease of the current efficiency for the reactions of electrolyte decomposition (6,7) can be explained only by an assumption that the F-P effect (of the type B) is connected with a partial recombination of deuterium

and oxygen. Indeed, the releasing-on-a-cathode deuterium can get onto an anode and be adsorbed on it.

Then, the adsorbed deuterium can take part in the thermal runaway reaction (8), i.e.



In a similar way also, the releasing-on-an-anode oxygen can get onto the cathode and be adsorbed on it. Then the adsorbed oxygen can take part in the symmetrical electrochemical reaction, i.e.



The overall reaction looks like as follows:



It should be noted that based on many-years experiments in their papers, Fleischmann and Pons themselves point out that for obtaining of the excess power effect (of the type B), one should seek conditions, where on an anode alongside with the reaction (7) in parallel, the electrochemical reaction (12) takes place; so they provide with this reaction in the paper [13] (in the Appendix 6).

For increasing of a cell effectiveness in producing of the excess energy, the authors [13] also provide with three recommendations.

First, to achieve for D_2 ionization at the anode, i.e. to achieve the occurrence of an electrochemical reaction (12).

Secondly, to make as little interelectrode gap as possible. In our proposed mechanism of the F-P effect (of type B) (12-14), this recommendation facilitates a penetration of deuterium onto an anode and of oxygen onto a cathode. So this way it is possible to increase intensity of the electrochemical reactions (12,13).

Thirdly, to increase an electrolyte concentration. This recommendation decreases a resistance and subsequently it results in a decrease of an electric potential and of an input power of the cell (5). Hence this leads to increase of cell's specific excess power P_{ex}/P_{in} . Thus, in our proposed mechanism of the F-P effect (of type B) (12-14), these recommendations increase an intensity of the parallel

It is interesting to note that if instead of palladium cathode to use a platinum cathode or if to use the palladium cathode not more than only one day, the F-P effect (of the type B) (or effect of decreasing of current efficiency) does not take place. Hence, the palladium (or more exactly the deuteride of the palladium, as the effect arises only after forming of a β -phase of the palladium deuteride) is a catalyst of the electrochemical reaction (13). The fact that the palladium cathode is a necessary condition of the F-P effect emergence is mentioned also in a lot of other papers [7-11,13].

With taking into account the possible parallel run of the three electrochemical reactions (6,7), (8,9) and (12,13), a cell power output can be written in the form:

$$P_{out}=(U-UI_m)i_1+442400(i_2/F)+(U-U2_m)i_2+(U-UI_m)i_3+UI_m i_3, \quad (15)$$

$$I= i_1+i_2+i_3, \quad (16)$$

where i_1 , i_2 , i_3 are current values in the parallel electrochemical reactions (6,7), (8,9) and (12,13) respectively. $UI_m=1.54V$ – thermoneutral voltage for electrolyte decomposition reactions (6,7) (Section 2) [13]; $U2_m$ - thermoneutral voltage for reactions of thermal runaway (8,9); F - Faraday constant. The first, third and fourth addends in the formula (15) determine the power output of Joule's heat released in a cell by a currents i_1 , i_2 , and i_3 respectively. The second addend determines the output power of exothermic thermal runaway reactions (8,9), where $|\Delta H_h|=442.4$ kJ/mol(D_2) is enthalpy of recombination reaction of the atomic deuterium [27]. The fifth addend determines the output power released in the cell on expense of the deuterium and oxygen recombination (exothermic reactions (12,13)). Indeed, $UI_m=|\Delta H|/2F \approx 1.54V$ (12,13), where $|\Delta H|=295$ kJ/mol(D_2) is enthalpy of the deuterium and oxygen recombination [13].

For the excess power from the equation (15) with due account of (5) and (16), we'll obtain the following equation:

$$P_{ex}=442400(i_2/F)+(UI_m-U2_m)i_2+ UI_m i_3, \quad (17)$$

As in whole over the entire period of the F-P effect (of the type B), the ratio between the deuterium concentration and the oxygen concentration makes 2:1, the role of the thermal runaway reactions (8,9) is insignificant (i.e. $i_2=0$). Consequently the excess power for this effect will be

$$P_{ex}=UI_{mi_3}, \quad (18)$$

A current value in the electrochemical reactions (12,13) can be found (using Figure 4(b)) from the equation:

$$i_3/I \approx (1-k_3), \quad (19)$$

where $k_3=0.815$ is a current efficiency for reactions of electrolyte decomposition (6,7) (Figure 4(b)).

Using the formulas (18,19) and the data from the Table 1 (experiment #107), we find:

$$I=0.2A, \quad i_3=0.037A, \quad P_{ex}=0.057W, \quad (P_{ex}/P_{in})100\%=6.3\% . \quad (20)$$

So the found theoretical value of the excess power P_{ex} (20) for the experiment #107 coincides with the experimental value obtained from calorimetric measurements (Table 1) within limits of relative error 1.8%.

These theoretical and experimental investigations prove categorically that the F-P effect (of the type B) is due to the partial deuterium and oxygen recombination in line with electrochemical mechanism (12,13). Thus, the F-P effect (of the type B) takes place on expense of energy of an external power source. At first an energy of the external power source is spent on decomposition of the heavy water onto deuterium and oxygen and then a partial recombination of the obtained deuterium and oxygen runs in line with the electrochemical mechanism (12,13). As a result, to the cell in the form of heat, a part is returned of energy spent on decomposition of the electrolyte. Hence the excess power (18) is apparent or imaginary.

For the experiment #65 (Table 1) for the current efficiency, we obtained the following experimental value:

$$k_3=94\% . \quad (21)$$

Using the value (21) and the formulas (18,19), one can verify that also in the experiment #65 the theoretical value of the excess power coincides with the experimental value obtained from calorimetric

We would like to attract your attention to the fact that in the experiments conducted by us (Table 1), the current efficiency values vary very much depending on intensity of the F-P effect (of the type B). This is why it is not surprising that in various works investigating the F-P effect, various authors inform on different values for this parameter in a range from values close to 100% [3,13] down to values about 80% and lower [2,40].

3.7 Reproducibility of the F-P effect

In the paper [11] based on a lot of investigations of the F-P effect, there was made the conclusion: “Notwithstanding that hundreds of effect explanation attempts have been made, there was not shown in them, how to improve an effect reproducibility and energy output.”

Only by establishing the true mechanism of the F-P effect can its reliable reproducibility be achieved. With this purpose, it is needed to exclude all occasional factors influencing the effect. Firstly, they are factors connected with electrodes construction. Secondly, they are factors connected with a moment of F-P effect initiation.

In investigations (Section 3.4), it was shown, that two processes of accumulation bring a cell step-by-step to the F-P effect. Firstly, this is the process of deuterium accumulating in electrodes. Secondly, this is the process of deposits accumulating on a cathode surface. Only in a case of a reliable realization of these two accumulation processes, the F-P effect is possible.

A mass of the deuterium accumulated in electrodes depends much on a number of micro-defects in the electrodes (micro-fractures, dislocations and suchlike) (Section 3.3). Notably that the larger number of the micro-defects in electrodes is, the more able they are to accumulate deuterium up to ceiling values pointed in the Table 2. In its turn, the number of the micro-defects depends on electrodes producer, a technology of their production and even of an electrodes commercial lot. Besides, the number of the micro-defects in electrodes is accumulated in the course of their operation [33,34]. In order to exclude all these occasional factors, from the very beginning, it is needed to use

electrodes with a very significant number of the micro-defects. The easiest way of doing so is a use of ceramic-metal Pd and Pt electrodes. Our many-years investigations [14-19] (and investigations in Table 2) of the process of accumulation of hydrogen/deuterium in electrodes showed that a use of the ceramic-metal electrodes is a necessary condition for the accumulation in electrodes of enormously great mass of hydrogen/deuterium [30,31].

So use of the ceramic-metal electrodes for the reproducibility of the F-P effect excludes occasional factors connected with electrodes manufacturing at all.

As it was shown earlier, a deposits density on a cathode surface is very important for the F-P effect emergence, as deposits are activation centers of the exothermic reactions of the thermal runaway (8,9), which do ensure the F-P effect. In order to exclude occasional factors connected with intensive deposits forming, it is enough to add palladium salts to electrolyte (it is possible to add nickel salts as it was performed in the paper [10]). As it is known, co-deposition of palladium in a course of heavy water electrolysis enhances a likelihood of the F-P effect emergence [36].

Completely these recommendations exclude occasional factors connected with electrodes preparation for the F-P effect obtaining.

To apply control to the power of the released energy resulted from the F-P effect is necessary to regulating mass of deuterium accumulated in electrodes. For this, it is necessary to accumulate the deuterium in the electrodes under a rather small current density, in order not to cause the F-P effect accidentally before a desired time moment. Usually we accumulated the deuterium in ceramic-metal Pt and Pd electrodes under the current density not more than 40 mA cm^{-2} during 100 days.

In accordance with the recommendations by F-P, an interelectrode gap is needed to be made as small as possible [13]. This will allow deuterium effective accumulation both on a cathode and on an anode, which is a necessary condition of the reactions running (8,9). The best of all is to use fabrics as separators between the electrodes. The fabrics can be the same that for the same purpose are used in alkaline batteries.

A beginning of the F-P effect is connected with the following occasional factor. Based on the conducted investigations, it is clear that for an initiation of the exothermic reactions (8,9) ensuring the F-P effect, it is to heat up intensively being-on-a-cathode active points formed by deposits. This can be done by letting a powerful current impulse passing through the cell, sufficient to decompose the deuterides accumulated in the electrodes (11), which will lead to the initiation of exothermic reactions (8.9). The current impulse value depends on constructive features of the cell (and first of all, on interelectrode gap) and deposits density on the cathode. In our experiments for the F-P effect initiation, usually, we used the current impulse ensuring the electrical potential on the cell more than 40 V during 0.25-0.5 s.

If to use all the pointed out recommendations, the F-P effect reproducibility will be obligatory.

It should be especially noted that the current of the F-P effect initiation must not be too high. In a case of really great heating of the cathode up, all the energy accumulated in it in the form of deuterides can be released not during 5.5 hours as it is shown in the Figure 3(a) but instead during several seconds. This will result in an explosion of the cell. The phenomenon of cells explosion is very rare and yet it was observed by a number of investigators and described in the papers [1,41-44]. As for this phenomenon, repeatedly we reproduced it in our laboratory, letting a current impulse through the cell, three times stronger than the one described above.

4. Conclusion

Based on the conducted investigations and data from the Table 1, it follows that in the process of a long-lasting heavy water electrolysis, inside of a cathode, the energy is accumulated in the form of the palladium deuterides with density equal to 0.381 MJ cm^{-3} (Table 1, #71, type A). This estimation is more than ten times less than estimation made by Fleischmann and Pons for specific energy released by a cathode, which was obtained based on erroneous account of the apparent excess power (of the type B) [1,13] and 5.036 MJ cm^{-3} (Table 1, #107). Nevertheless, the obtained by us real specific energy being accumulated by a cathode exceeds significantly the specific energy of contemporary

organic energy carriers [45], which opens great possibilities for development of new forms of power industry.

Undoubtedly, the F-P effect requires further both experimental and theoretical investigations. Nevertheless, the determined in this paper mechanism of the F-P effect allows explaining all available currently experimental data.

Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

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Table 1

Parameters of Fleischmann-Pons effects in different experiments.

Experiment number	71	71	65	107
Type of F-P effect	A	B	B	B
Diameter × cathode length (cm)	0.4×1.25	0.4×1.25	0.4×1.25	0.2×1.25
Current density (mA cm ⁻²)	64	64	64	256
Cell average voltage (V)	3.50	3.74	3.76	6.03
F-P effect duration (h)	5.49	912.4	945.2	974.3
Input power (W)	0.198	0.22	0.222	0.898
Excess power (W)	3.029	0.01	0.0094	0.056
Excess energy (J)	59873	32846	31986	196419
Power output/Input power, avg [max]	16.3[41.2]	1.045	1.042	1.062
Specific excess power (W cm ⁻³)	19.3	0.064	0.060	1.436
Specific excess energy (MJ cm ⁻³)	0.381	0.209	0.204	5.036

Table 2

Analysis results of deuterium accumulating in electrodes of Fleischmann-Pons cell.

Electrode number	71	71	54	54	107	107
Electrode type	Pd	Pt	Pd	Pt	Pd	Pt
Electrode Diameter×Length (cm)	0.4×1.25	0.1×15	0.4×1.25	0.1×15	0.2×1.25	0.1×9.5
Deuterium volume (l)	2.525	1.311	0.198	0.103	0.063	0.084
Electrode weight (g)	1.810	2.501	1.889	2.527	0.472	1.600
Gravimetric capacity (wt%)	19.9	8.6	1.8	0.7	2.3	0.9

Fig. 1. Change in parameters of the KSX-25 battery during thermal runaway: I is the charging current of the battery, U is the voltage at the battery terminals: a) battery No. 1, b) battery No. 2.

Fig. 2. a) Oxide–nickel electrode of KSX-25 battery (No. 1) and b) cadmium electrode of KSX-25 battery (No. 2) after thermal runaway. Dimensions of the electrodes equal to 7.3×13.6 cm. Black circles are the places where the separator burned out.

Fig. 3. Changes of cell parameters during Fleischmann-Pons effect (of type A). a) Change of ratio (power output)/(input power) versus time. b) Change of cell voltage versus time. Cathode 0.4×1.25 cm Pd rod, electrolyte 0.1 M LiOD solution, current density 64 mA cm^{-2} , bath temperature 30°C .

Fig. 4. Changes of cell parameters during Fleischmann-Pons effect (of type B). a) Change of ratio (power output)/(input power) versus time. b) Change of averaged-out (with interval equal to 7 hours) efficiency of the heavy water decomposition current onto deuterium and oxygen versus time. Cathode 0.2×1.25 cm Pd rod, electrolyte 0.1 M LiOD solution, current density 256 mA cm^{-2} , bath temperature 30°C .

Contributions of all authors are the same. All parts of the work (on the manuscript) were carried out jointly.

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Declaration of interests

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

The authors declare the following financial interests/personal relationships which may be considered as potential competing interests:

Journal Pre-proof

- *Effect of Fleischmann-Pons is not associated with cold fusion of deuterium nuclei.*
- *Effect of Fleischmann-Pons is associated with exothermic reaction of thermal runaway.*
- *During of heavy water electrolysis deuterium accumulates in the electrodes.*
- *Recombination of accumulated atomic deuterium is the reaction of thermal runaway.*
- *Recommendations are given allowing reproducing this effect without failures.*

Journal Pre-proof

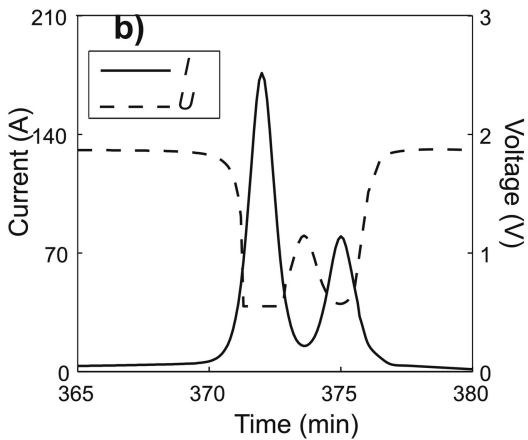
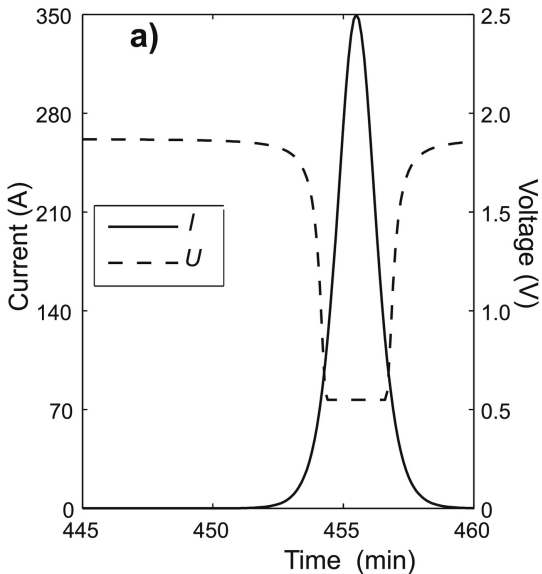
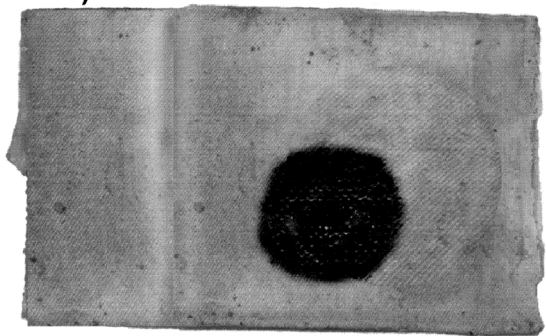


Figure 1

a)



b)

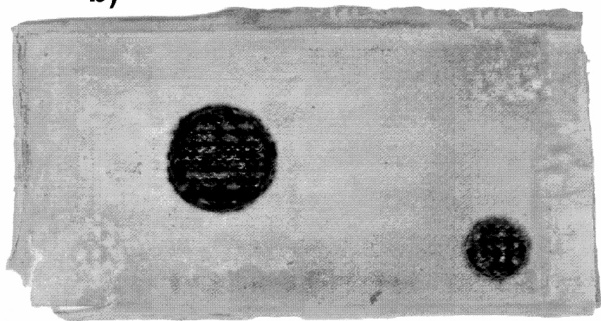


Figure 2

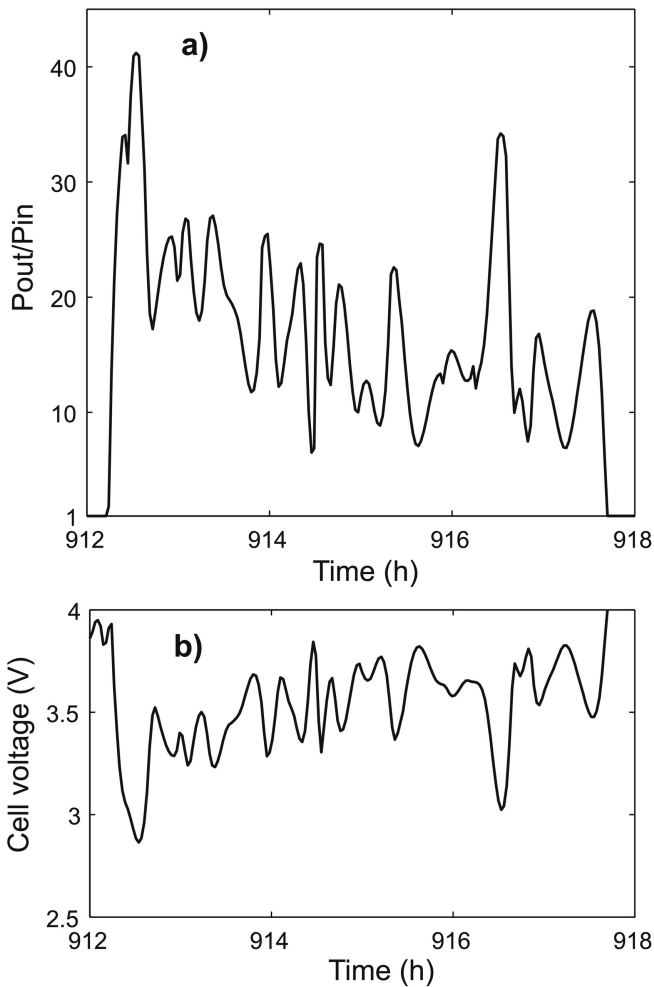


Figure 3

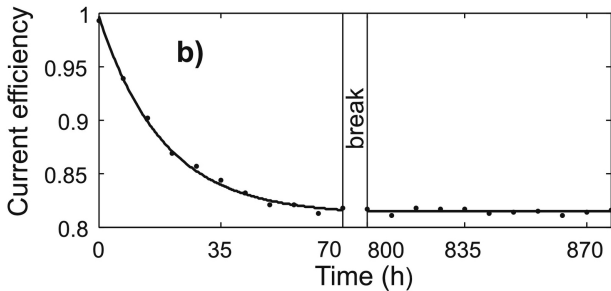
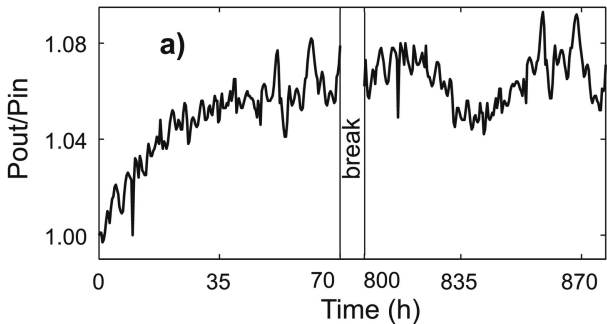


Figure 4