# The Most Extensive Measurement of Excess Energy by M.C.N. McKubre et al. (1993, 1994)\*

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The measurement of excess energy, one of the indirect evidences of the cold fusion phenomenon, was the most reliable result among others in the content of the pioneering paper by Fleischmann et al. published in 1989 [Fleishmann 1989]. They used PdD<sub>x</sub>, one of the typical cold fusion materials (CF materials) of the deuterium system. (By the way, we would like to recall that another CF material used frequently in the CFP research is NiH<sub>x</sub> as discussed in the previous article of the "*From the History of CF Research* (3)" (*CFRL News* No. 89)).

The experimental data sets published by McKubre et al. in 1993 and 1994 in the same  $PdD_x$  system as that by Fleischmann et al. with a more sophisticated apparatus are the most extensive one ever obtained [McKubre 1993, 1994a, 1994b]. For convenience of the readers, we posted the first two papers at the CFRL website after the CFRL News No. 91. We investigate the elaborate result by McKubre et al. in this paper and elucidate its indispensable value for the science of the cold fusion phenomenon (CFP).

# 1. Characteristics of the Experiments by McKubre et al. [McKubre 1993, 1994a, 1994b]

The extensive work on the excess energy generation by McKubre et al. had been published in this and following several papers [McKubre 1993, 1994a, 1994b]. In this article we focus mainly on the first paper presented at ICCF3 to show that the extensive data sets obtained by a sophisticated apparatus give us much useful information about the physics of the cold fusion phenomenon (CFP). We posted the first two papers by McKubre et al. at this News site after this CFRL News No. 91. Their experiment was prepared to check the possible nuclear reactions presumed by Fleischmann et al. [Fleischmann 1989] using a state-of-the-art apparatus they constructed for a precision measurement of excess energy as expressed in their paper ad follows:

"Thus a central feature of the experiments described here is the (electrochemical) control, and continual in situ measurement, of the deuterium (and hydrogen) loading during the entire calorimetric experiment. Further, it was decided that the most accurate, and sensitive, thermal measurements would be obtained using a sealed (thermodynamically closed) electrochemical cell (with knowledge at all times of the composition of the reacting system), in combination with a flow calorimeter." ([McKubre 1994a] 1. Introduction).

They used a sample of 0.1 cm diameter  $\times$  45 cm length Pd cathode and Pt wire anode in an electrolyte 1.0 M LiOD + 200 ppm Al in D<sub>2</sub>O. They obtained an abundant experimental data in a time interval 300 – 780 h after the start of the experiment as shown in Fig. 1 and 2 (Figs. 6 and 7 of [McKubre 1993], respectively).



Fig. 1 Variation of excess power with cell current (Fig. 6 of [McKubre 1993])



Fig. 2 Variation of excess power with loading (Fig. 7 of [McKubre 1993])

Figure 1 shows the observed excess power P(i) (W) as a function of the cell current *i* (A). The excess power P(i) appears for the cell current *i* larger than a threshold value  $i_0$  ( $i_0 \sim 0.5$  A) and the average value  $\langle P(i) \rangle$  of P(i) increases with the current as ( $i - i_0$ )<sup> $\alpha$ </sup> ( $\alpha \sim 1$ ) with a very large dispersion.

Figure 2 shows the observed excess power P(x) (W) as a function of the average loading ratio  $x \equiv D/Pd$ . The average loading ratio x was determined by the known resistance-loading variations data of the cathode. The excess power P(x) appears for the loading ratio x larger than a threshold value  $x_0$  ( $x_0 \sim 0.85$ ) and the average value  $\langle P(x) \rangle$  of P(x) increases with the current as  $(x - x_0)^{\beta}$  ( $\beta \sim 2$ ) with a very large dispersion as in the case of Fig.1.

Figures 1 and 2 show clearly that the excess power P obeys a law in its average but with a very large dispersion around it. The reliability of the measurement is widely recognized despite of the large dispersion of the data as we see in the DOE report 2004 as follows [DOE 2004]:

"Electrolytic experiments are extremely difficult to conduct properly and are not geometrically compatible with many detectors for radiation or nuclear particles. This explains the shift to non-electrochemical approaches, which should continue. Emphasis should be placed on developing theories that explain existing data and guide future experimental work. New experiments that test the underlying principles of the theory should be performed. The body of work that has resulted from LENR investigations is formidable and worthy of attention of the broader scientific community." ([DOE 2004], Comment #9)



Fig.3 Variation of excess power, uncertainty and loading (Fig. 5 of [McKubre 1993])

In addition to the dependences of the excess power P on the cell current i and the loading ratio x given in Figs. 1 and 2, respectively, they observed dynamical behavior of the experimental system as shown in Fig. 3 (Fig. 5 of [McKubre 1993]). In this figure, we see that the excess power P and the loading ratio x had shown complex temporal variation. This complex behavior may be an evidence of some fundamental nature of the events in the CFP as discussed in Section 1-3.

## 2. Behavior of Average Values of Excess Power

According to the data and suggestions given in the original paper, we can deduce an empirical relation between average values  $\langle P(i, x) \rangle$  of the excess power *P*, the loading ratio *x*, the current density *i* and the loading speed d*x*/d*t* as follows:

 $\langle P(i, x) \rangle = M(T, /dx/dt/)(i - i_0)^{\alpha} (x - x_0)^{\beta},$  (1)

where M(T, |dx/dt|) is a constant depending on the temperature T, |dx/dt|, quality of the Pd cathode, the surface condition and others,  $i_0$  and  $x_0$  are threshold values of i and x, respectively, and  $\alpha$  and  $\beta$  are constants with values  $\alpha \sim 1$  and  $\beta \sim 2$ , respectively. The threshold values of the current density i and the loading ratio x are taken as  $i_0 \sim 0.2$  mA/cm<sup>2</sup> and  $x_0 \sim 0.85$ .

The dependence of the excess power P on the dx/dt is concluded somewhat arbitrarily from their conclusion "iii) changes in the excess power level are usually associated with departures from the electrochemical steady-state, caused primarily by varying the current." On the other hand, it is also concluded that "the maintenance of high loading for considerable period of time" is required for excess power generation [McKubre 1994a, Subsection 4]. By this comment, the dependence of P on the variation of x (or i) may be spurious.

It should be kept in mind that the variables in this equation are not independent each other, i.e.  $i_0$  is dependent on the variable x and  $x_0$  is dependent on i, and so on.

About the constant M(T), we have now an impression that it increases with temperature as several experimental data done since 1995 show it (e.g. [Kozima 2007]).

### 3. Dispersion of the Data Points

In the previous subsection, we deduced an empirical formula between the average values of the excess power P and the loading ratio x and the current density i even if we know the large dispersion of the data points in their Figs. 1 and 2 (Figs. 6 and 7 of [McKubre 1993], respectively). The dispersion of the data points for a fixed value of x or i appeared in these figures is more directly expressed in their next paper as follows: "It is worth noting, however, that excess power in these four experiments (P13 – P16) was not produced in exactly the same amounts, or at exactly the same times, in response to the same stimuli." ([McKubre 1994a] Section 4).

#### 4. Analyses of the Data by McKubre et al.

From our knowledge of nuclear transmutations in the CFP, the nuclear products observed by now localize at surface regions within several micrometers. This fact has been taken into our model (the TNCF model) where reactions of trapped neutrons with isolated nuclei occur in the surface layer having a width of about several micrometers  $l_0$ , which depends on the experimental condition.

It is interesting to note that the surface layer of the cathodes used in their experiment has attracted their notice as follows; "This observation raises the interesting possibility that one or more species, other than deuterium, are required to be present in the cathode in order to observe excess power, species which are not present initially and are thus required to diffuse into the cathode, presumably from the electrolyte. Analyses of used cathodes have revealed the presence of several light elements in the near-surface region (to a depth of several microns); in particular, lithium." ([McKubre 1993], Section *Phenomenological model*). This description is in accord with our suppositions made in

the TNCF model.

#### 4-1 Average Behavior of Excess Energy Production

We have given an interpretation of the meaning of Eq. (1) in our previous paper [Kozima 1996] based on our TNCF model developed until that time and explained also in our book [Kozima 1998].

It should be kept in mind that the factor M(T, |dx/dt|) in Eq. (1) should be written as  $M(T, |dx/dt|, \zeta)$  where the parameter  $\zeta$  represents variables in the system other than temperature and |dx/dt| to express the dependence of the excess power generation on the parameters of the system while the equation was analyzed in a somewhat different form in the previous treatment [Kozima 1996, 1998].

The Eq. (1) should be reanalyzed from our novel point of view of the trapped neutrons developed recently and be presented elsewhere.

# **4-2** Wide Dispersion and Temporal Variation of the Experimental Data around its Average – Complexity

Then, our next question is what the dispersion means in the cold fusion phenomenon as seen in Figs. 1 and 2 and explained in [McKubre 1994a] and what the temporal variation as seen in Fig. 3.

We have to recognize the situation where the  $PdD_x$  samples are fabricated in the experiment where the measurements of excess power had executed. In each process resulting in the formation of the  $PdD_x$  cathode (a CF material), there are many microscopic processes of atoms and ions; occlusion of deuterium into the Pd cathode from electrolyte ( $D_2O + LiOD$ ), formation of the PdLi<sub>x</sub> surface layer over the surface of the cathode, migration of electrons and deuterons in the cathode accelerated by the application of electric bias and so on. We have to accept the inhomogeneous distribution of deuterons in the cathode especially along the radius of the wire and self-organization of a stable structure in a localized region at the surface of a sample

These factors existing in the experimental conditions induce complexity well known in the nonlocal dynamics. Therefore, we have to take into our consideration the statistical point of view to investigate the elaborate experimental data obtained by McKubre et al. if we want to understand the data as a whole.

One of such trials had been given in our paper where the data given in Fig. 2 (Fig 7 of [McKubre 1993]) to deduce an empirical law "inverse-power dependence of frequency on intensity of excess heat production" [Kozima 2006 (Sec. 2.12), 2012 (Sec. 2.2)]. The data shown in Fig. 3 (Fig.5 of [McKubre 1993]) was used to relate the CFP

with the chaotic behavior of nuclear reactions in such cold fusion materials (CF materials) as  $PdD_x$  [Kozima 2012, Sec. 2.3].

#### 5. Conclusion

The extensive experiments by McKubre et al. [McKubre 1993, 1994a, 1994b] have revealed various important features of the CFP from the dependence of the average excess power P on the cell current i and the loading ratio x to complex behavior (complexity) of the events of the excess power generation. The latter behavior should be taken as a fundamental but not accidental or artifactual feature of the CFP which resolves the problem of absence of the quantitative reproducibility (or rather existence of the qualitative reproducibility) of the experimental results as already shown clearly in the paper by McKubre et al. [McKubre 1994a] cited above in Subsection 1.3.

The fact that they did not observe occurrence of the CFP in their protium system PdH<sub>x</sub> (P13 of [McKubre 1994a]) is not in contradiction to our mechanism of the CFP between trapped neutrons and nuclei in the surface region where the cf-matter is formed [Kozima 2006 (Sec. 3.7), 2014 (Sec. 5)]. In a CF material PdD<sub>x</sub> at ordinary temperature, in their case at 30 °C, a deuteron has rather wide-spread wavefunction than a proton while the situation reversed at higher temperature [Kozima 2014, Appendix A3]. It is possible, therefore, to expect nuclear reactions in PdH<sub>x</sub> at higher temperatures which could be the cases of the episode of Pons (cf. *CFRL News* No. 89, Sec. 1) and of the data obtained by Jones et al. (cf. *CFRL News* No. 90, Sec. 1.2).

Using the elaborate data obtained by McKubre et al. [McKubre 1993, 1994a, 1994b] together with other data of nuclear transmutation and neutron emission in the CFP, we can figure out the physics of the cold fusion phenomenon as that primitively describe by our TNCF model and can understand the importance of the behavior of neutrons in CF materials.

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