

## Concerning Reproducibility of Excess Power Production

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### **Abstract**

An apparent irreproducibility in the production of an, as yet, anomalous excess power from Pd cathodes electrochemically loaded with D can be associated with irreproducibility in the attainment of several necessary starting conditions. Of these, the threshold loading (D/Pd atomic ratio) has received the most attention. A statistical analysis is presented of the results of 176 experiments intended to test the means of establishing reproducible control over D/Pd loading. A set of variables are examined, and procedures identified which permit the attainment of loading above the threshold necessary for excess heat production.

Calorimetric results from two experiments are presented and analyzed. A mathematical function is identified which correlates closely with the time evolution of excess power. An important element of this correlation is the measured rate of change of the cathode resistivity. We have interpreted the resistance change as indicating the presence of an oscillation or "breathing" of the cathode loading induced by a flux of deuterons through the cathode/electrolyte interface.

The observed functionality of excess power with deuteron flux above a loading threshold, conforms closely with theoretical predictions.

### **1. Introduction**

Six years have passed since publication of the defining paper of Fleischmann, Pons and Hawkins.<sup>1</sup> In this time, scientific progress has been made to resolve issues that were quickly, and easily, anticipated. The resolution of some important issues has been accompanied by a slow, somewhat unsteady, but certain growth in the field of science which has come to be called "cold fusion".

At the time of that publication, few would have suspected after six years of study, that the field revealed would have survived, but not thrived. This surprising situation has come about because, in restraint of progress, stand two technically challenging problems:

- i) Irreproducibility. This is not, as some have argued, an irreproducibility of results. It is a difficulty in achieving reproducible starting conditions for comparative experiments.
- ii) Scarcity of energetic (nuclear) products. It is quite clear that the nuclear product(s) quantitatively associated with excess heat, if there is one, is not an energetic particle, or penetrating radiation, or a radioactive isotope; this makes the search very difficult.

In this paper, an attempt is made to address the first of these difficulties, with specific attention to the reproducible attainment of positive heat excess in the D/Pd system, under electrochemical conditions.

## 2. Necessary Conditions

We need first to identify the experimental starting condition thought, or observed, to be associated the phenomenon under study. In a series of papers SRI<sup>2-7</sup> and others<sup>8,9</sup> have attempted to define and quantify the variables associated with apparent excess heat production. These are:

i) Loading. The D/Pd loading seemed, at the outset, to be a likely controlling variable; this has clearly been shown to be the case. The loading is relatively easily measured; the attainment and maintenance of high loading ( $D/Pd \geq 0.9$ ), is not easily controlled. Independent experiments at SRI<sup>4</sup> and IMRA-Japan<sup>8</sup> have demonstrated a roughly parabolic functionality between excess power and loading above a loading threshold of  $D/Pd \sim 0.84 \pm 0.02$ . In order to achieve appreciable (and measurable) power excess, loadings much higher than the threshold are needed ( $D/Pd \geq 0.9$  and preferably  $\geq 0.95$ ). Several other factors also are necessary.

ii) Current Density. In an electrochemical experiment, current density affects loading. In addition to this expected, but complex, functionality, the interfacial current density also directly affects the rate of excess heat production. In experiments performed under nearly isothermal conditions at nearly constant loading, a linear dependence of excess power is observed with increasing current density above the threshold value. Unlike the loading threshold, the current density threshold displays considerable variability ranging from  $\sim 100$  to  $\sim 400$  mA cm<sup>-2</sup> in different experiments, or in the same experiments, at different times.

iii) Initiation. Many authors have reported a delay in the onset of excess heat production. Even after the loading and current density thresholds have been usefully exceeded, excess heat is (generally) not observed until a significant time has elapsed, on the order of 300 h for 1-4 mm dia. Pd wires. It is apparent that some initiation process must occur, presumably within the Pd lattice. At this stage, the origin is unclear.

iv) Disequilibrium. It has long been suspected that a flux of deuterium atoms through the interface is needed in addition to high deuterium atom loading, to produce measurable excess power. Recently theoretical arguments have been advanced to support the conjecture that flux, combined with high loading are necessary for excess heat or nuclear product formation. In this paper, we present experimental evidence of this, and attempt to quantify the role of deuterium interfacial flux in excess power production.

### 3. Loading

The attainment of high D/Pd loading is critical to the appearance of excess power. We previously have presented evidence<sup>2-7</sup> that, in every case that we have observed in a calorimeter, where a cathode achieved a maximum loading of  $D/Pd \geq 0.95$  while meeting criteria (ii) and (iii) above, excess power was measured ( $P_{xs} > 0$ ). For cathodes achieving maximum loadings  $0.90 \leq D/Pd < 0.95$ , approximately half exhibited  $P_{xs} > 0$ . In only one case for which the maximum loading was less than 0.90, have we observed  $P_{xs} > 0$ . This case will be discussed later in this paper, as it is instructive.

Attainment of these high loadings, while clearly critical, is neither usual, nor can it be achieved reproducibly. An analysis is presented here of the results of 214 experiments, both loading and calorimetric, representing 200,416 hours of experiment operation (or 23 experiment-years). All experiments employed Pd cathodes, and loading was measured using a four terminal resistance method.<sup>3,7,10</sup>

From this set of 214 experiments, the data from 38 have been excluded by reason of poor resistance measurement, premature failure, or suspected compromise, leaving 177,640 hours of reliable data, (20 experiment-years). The following Tables show the material sources, electrolytes and additives employed.

**Table 1. Metal Sources**

Engelhard					Various
E1	32*	Engelhard Lot #1	AE	1	AECL**
E2	24	Engelhard Lot #2	AM	4	Aithaca Metals
E3	38	Engelhard Lot #3	IB	4	Ingot Block - cast
E4	4	Engelhard Lot #4	JM	40	Johnson Matthey
E5	15	Engelhard Lot #5	NL	5	NRL†
E-	6	Engelhard 1mm	ZR	1	Zone Refined

\* Number of experiments

\*\* AECL Atomic Energy of Canada Ltd (Cast, High Purity)

†NRL Naval Research Laboratories (High Purity). 30 and 600  $\mu$  grain sizes.

**Table 2. Electrolytes, Additives and Variables**

	Electrolytes		Additives††		Variables
3	0.5 D <sub>2</sub> SO <sub>4</sub>	D	71	None	Additive
6	0.5 Li <sub>2</sub> SO <sub>4</sub>	D	68	Al	Annealing
158	1.0 LiOD	D	10	Ni†††	Cathode machining
2	0.3 M LiCl	D	6	B	Diameter
1	0.1 M LiOD	D	6	Cu	Experiment length
1	0.5 Li <sub>2</sub> SO <sub>4</sub>	H	6	Poison*	Metal source
4	1.0 LiOH	H	3	Si**	
1	0.1 M LiOH	H	2	Be	

\* Additional series of experiments were performed specifically to test the effects of classical hydrogen recombination poisons and the effect of their concentrations. The results of these have not been included in this data set.

\*\* All experiments were performed in quartz vessels and/or with quartz structures in the electrolyte. Si, was therefore present in increasing amounts as this element dissolved. For these three cases, additional Si was added as SiO<sub>2</sub> dissolved in electrolyte.

†† For the 176 cases analyzed here, when present, additive concentrations were ~ 200 ppm.

††† These 10 experiments employed Ni anodes not additives.

Each experiment has been characterized by the **maximum loading** attained by the cathode, at any time during the experiment (the average experiment length was 1009 hours). Twelve approximately equal groups were defined, according to the mean loadings shown in Table 3. An exhaustive analysis of these results is presented elsewhere.<sup>11</sup> The purpose here is to focus attention on those variables that strongly influence loading; this we can do with a relatively simple analysis.

The results in Table 3 show great dispersion, with a significant number of experiments achieving a maximum loading of only 0.74 (just beyond the maximum in R/R<sup>o</sup>), while an equal number apparently exceed 1.0 (R/R<sup>o</sup> ≤ 1.5 on the right hand side of the maximum). At this point, it is worth remembering that the experiments analyzed here represent our "best efforts" to attain high loading; 57% of all experiments reported in Table 3 achieved our stated target of 0.9.

**Table 3. Maximum Loading**

Group	Mean	#	Quartile	Group	Mean	#	Quartile
1	0.74	14	4	7	0.92	13	2
2	0.75	13	4	8	0.93	1	2
3	0.83	18	4	9	0.94	15	2
4	0.87	14	3	10	0.95	13	1
5	0.89	13	3	11	0.96	16	1
6	0.90	16	3	12	1.02	14	1

We are interested in the variables, of those examined, which affect the attainment of high loading. We have divided the results into quartiles. Very roughly, we are interested in the correlation between the experimental variables, and attainment of the first (preferably) or first and second quartiles. In Table 4 we indicate the variable, the number of experiments in that set, the percentage attaining the first quartile, and the percentage attaining the first or second quartile.

**Table 4. Variables Affecting Loading Quartiles**

Pd Lot Variation:	#	First	First or Second
E1	32	53%	78%
E2	24	29%	67%
E3	38	26%	42%
Not {E1,E2,E3}	82	12%	38%
E5	15	13%	80%
JM	40	18%	35%
<b>Additives:</b>			
Al	68	34%	57%
Ni	10	0%	0%
None	71	17%	48%
<b>Physical Variables:</b>			
Machined	39	36%	77%
d<0.25 cm	42	14%	26%
t>1009 h	70	34%	63%

Table 4 reveals a striking variation between Pd samples, even in lots from the same manufacturer. For Engelhard Lot#1 (E1), 53 % loaded into the first quartile while 78 % reached the first two quartiles. Whatever property this material had (or, critical flaw it did not have), there was less of it in subsequent lots, which exhibited generally inferior behavior.

For various samples from Johnson Matthey (JM), the pattern is not so clear. On average, these materials performed poorly (although better than other samples in the not {E1, E2, E3} set). The distribution for all JM samples is, however, bimodal, exhibiting a peak in Group 5 and another in Group 12.

For the effect of additives, only Al and Ni were sampled in statistically significant numbers. For Al, the effect is moderately strong and positive; for Ni (in this cases as an anode, not an additive) the effect is strong and deleterious. The set with no additives {None} loaded less well, on average, than those with deliberate additives.

In the final category, strong effects are seen for machining, cathode diameter, and experiment duration. "Machined" refers to those cathodes for which the outer surface was removed mechanically, by a cutting tool, in a lathe. In this process, the radius (of a 1/8" or 3mm diameter) cathode was reduced by ~ 0.1mm (respectively to 3mm or 2.8mm diameter), in a single pass of the cutting tool. This process appears to be beneficial in removing surface inclusions and mechanical defects, and in promoting loading.

In our experiments, smaller diameter wires ( $d < 0.25$  cm) have not loaded, on average, as well as cathode 2.8 mm in diameter and larger. It should be noted that none of the smaller diameter wires were machined prior to electrolysis.

The final variable, duration of experiment, needs to be interpreted with care. Experiments longer than average ( $t > 1009$  h) correlate with high loading. This reflects two factors. Cathodes which initially load poorly (fourth quartile) tend, not to respond with better loading following anodic stripping or chemical addition, and these experiments often were terminated early. Cathodes which initially load at least into the third quartile, often display increased loading after anodic stripping cycles with or without chemical addition. This improvement may continue in several sequential loading cycles, each of which may require 200-400 hours. Such experiments tend to be longer than average and to result in better than average loading.

#### **4. Metallurgical Variables in Loading**

It is clear from Table 4, that the metal, itself, plays a role in determining the maximum loading. One might propose, as a hypothesis, that a difficulty in attaining reproducible starting conditions, in this case threshold loading, is due in large part to an uncontrolled variable contained within the Pd stock. Anticipating this possibility, the 176 cathodes analyzed in this paper were all prepared from moderate to high purity Pd. All were subjected to a similar annealing process (800-850°C for 2-3 hours, in low O<sub>2</sub> Ar, vacuum, or D<sub>2</sub> gas). With only one or two exceptions, all were etched with aqua regia (appropriately light or heavy) after machining and annealing. These procedures were intended to assert a uniform bulk and surface condition on the cathodes; nevertheless, significant variability was encountered between and within Pd lots.

Several important questions are raised in this analysis:

- i) What is the characteristic of a "good" material?
- ii) What is the uncontrolled parameter?
- iii) How can we control this parameter to yield consistently high loading?
- iv) How can we maximize the loading for a "good" Pd sample?
- v) Does this procedure work to optimize loading in "poor" Pd samples?

We can begin to answer some of these questions by further analysis of our loading data base. Examining Table 4 we see that the parameters associated with the attainment of high loading are: Metal Source; Machining; {Annealing}; {Etching}; {1M LiOD} + Al addition. { } is a parameter employed, but not tested statistically. At this point, it is worth noting that a significant variable, the presence of dissolved silicate in solution, has also not been tested in this data set. Previous experiments<sup>12</sup> have indicated that Si is critical for the attainment of high loading. In all of the experiments described here this species was provided adventitiously by dissolution of quartz components of the electrochemical cell.

We can therefore propose a "best" procedure to achieve loading, based on the set of parameters examined. In this procedure one would:

- [ 0) *Select a suitable material* ]
- 1) Machine the outer surface to remove surface inclusions and damage.
  - 2) Anneal at low oxygen partial pressure, in the temperature range 800-850°C, for 2-3 hours.
  - 3) Etch cathode (plus Pt contact wires) in freshly prepared (heavy) aqua regia, for 5-10 seconds.
  - 4) Electrolyze in 1 M LiOD, freshly prepared (in a H<sub>2</sub>O free environment), by the reaction of D<sub>2</sub>O with high purity Li.
  - 5) After electrolyte preparation, and shortly before electrolysis, include in the electrolyte a small piece of high purity Al foil, sufficient to yield 200 ppm when dissolved.

The results are shown in Table 5 for experiments in which this enumerated procedure was followed.

	#	Quartiles	
		First	First or Second
<b>Procedures 1-4 (No additive)</b>			
E1	11	27%	55%
E2-4	6	33%	83%
Other	10	10%	80%
All	27	23%	69%
<b>Procedures 1-5 (Al additive)</b>			
E1	8	88%	100%
E2-4	4	100%	100%
Other	9	22%	78%
All	21	62%	90%
<b>Procedures 2-5 (No machine)</b>			
E2-4	26	23%	50%

While the selection constraints are severe, and the numbers in each category are small, a clear pattern emerges. Without the addition of Al, Procedures 1-4 show a small benefit for the Engelhard samples but no significant effect for other samples. In the presence of Al, however, following Procedures 1-5 yields a striking and useful result for Engelhard samples, and a beneficial effect for all metal samples.

We cannot generally assess the influence of machining as all E1 samples were machined before use. Procedures 2-5 (with Al, no machining), results in no significant improvement in maximum loading for the set of Engelhard samples {E2-4}.

From the analysis presented here, one may conclude that surface machining, and the addition of Al to the electrolyte both are important in concert, but that neither has a strong influence, alone. This concerted action, or synergism, benefits both the "good" loading materials, specifically early Engelhard Lots, and other generally more difficult to load Pd samples. In the light of this discovery, we must focus attention on the underlying mechanisms of loading, in order to reveal the hidden variable which influences the reproducibility of loading.

## **5. Other Issues of Reproducibility**

The D/Pd loading is clearly a critical element in attaining the conditions that have been shown to be necessary for excess heat production. In the past six years, the issue of loading, and its reproducible attainment for both the D/Pd and H/Pd systems, has been studied extensively and with renewed vigor. Our understanding, however, remains poor. Furthermore, other parameters have been shown to be critical for the reproducible attainment of excess power: interfacial current density; time delay or initiation. In neither case is the role of these variables, or the process(es) that they engender, understood. Either may contribute significantly to an (apparent) irreproducibility of effect.

Recent experimental results have indicated a significant mathematical correlation between the rate of excess heat production, and the rate of change of the Pd cathode resistance. This effect, if not monitored, may constitute another important factor in (apparent) irreproducibility. If monitored and understood this phenomenon may yield information about fundamental processes and the underlying mechanism of excess heat production.

We measure cathode resistance, using a four-terminal method<sup>3,7,10</sup>, to monitor D/Pd loading. Two parameters importantly control the resistance of palladium: hydrogen isotope loading, and temperature. In the experiments to be discussed, we observed a periodic variation in Pd resistance with fundamental period ~ 2 hours (~  $10^{-4}$  Hz), the amplitude of which appeared to correlate directly with the magnitude of the excess power being measured simultaneously in a mass flow calorimeter.<sup>6,7</sup>

If we accept that  $R/R^0$  and  $P_{xs}$  both are measured accurately, then, as an explanation for this correlation we offer two hypotheses:

i) The observed excess heat is sourced at one or more small regions within the cathode. Because of the small volumes, local heating of the cathode produces resistance fluctuations which are observed at effectively constant loading. In this hypothesis, the excess power is the "cause", and the resistance the "effect". Periodicity of the resistance (perhaps induced by local de-loading following local heating) should be

associated in this model with a periodicity in excess power with the same frequency. Such an effect is indeed, observed.

ii) One can propose an alternative hypothesis in which resistance fluctuations reflect the "cause" with excess power the "effect". Unless the source of excess heat is extremely localized, at the levels of average excess power observed ( $1-5 \text{ W cm}^{-3}$ ) one would expect changes in local temperature insufficient to cause a detectable resistance variation. In which case one must look to a change in loading to cause a change in resistance. Bulk loading changes by adsorption/desorption processes at the cathode/electrolyte interface; this is accompanied by a flux of deuterons orthogonal to, and through the interface. The rate of change of average loading ( $\delta x/\delta t$ ) is thus related to the deuteron flux.

There has been a suggestion<sup>13</sup> that, above a critical loading threshold, the rate of excess heat production should be related to the deuteron flux (*i.e.*  $\sim \delta x/\delta t$ ). For this reason we have chosen to analyze our data in terms of hypothesis (ii). It should, however, be remembered that an alternative hypothesis exists which is consistent with the experimental evidence so far obtained.

## 6. Results

Figure 1 presents the current density, loading and excess power, measured by the methods previously described,<sup>2-7</sup> for a 1 mm dia., 10 cm long Pd cathode. The palladium was obtained from Johnson Matthey, and was formed into the shape of a horizontal "lasso", and annealed by our normal procedure.<sup>7</sup> The electrolyte was 1.0M LiOD containing 200 ppm Al at the outset. Sufficient Cu was added, dissolved in LiOD, 156 hours before the data shown in Figure 1, to make the concentration  $\sim 3$  ppm in the electrolyte.

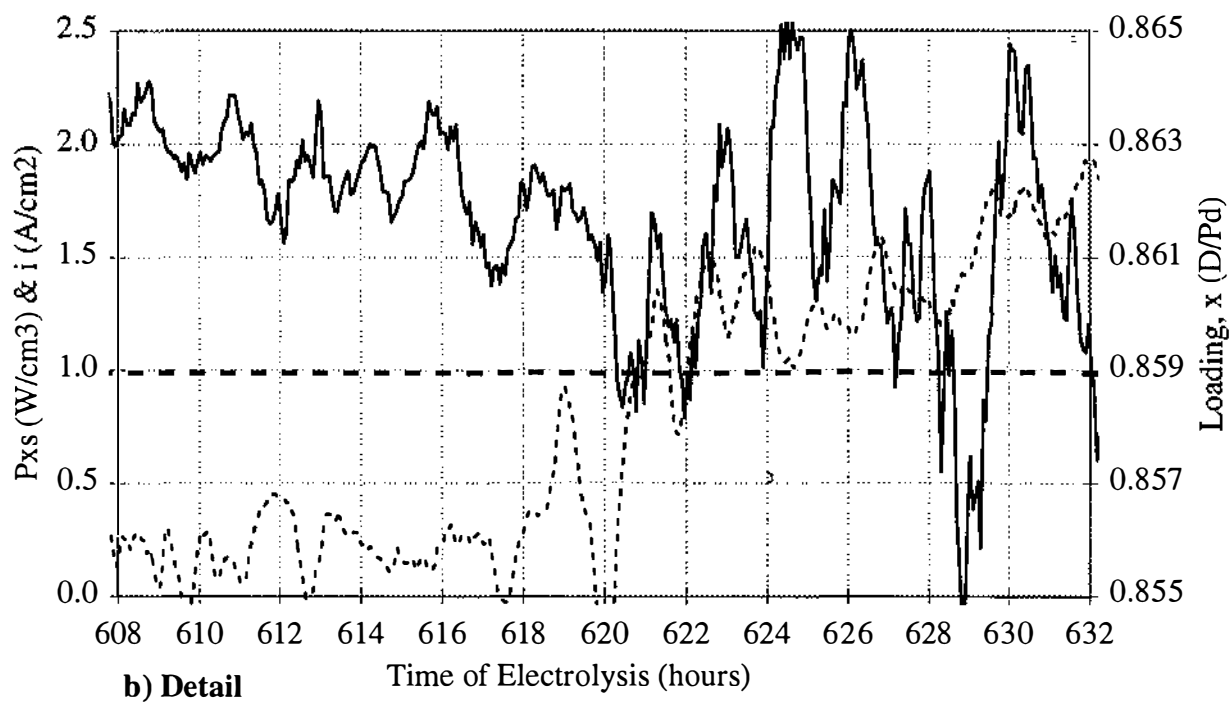
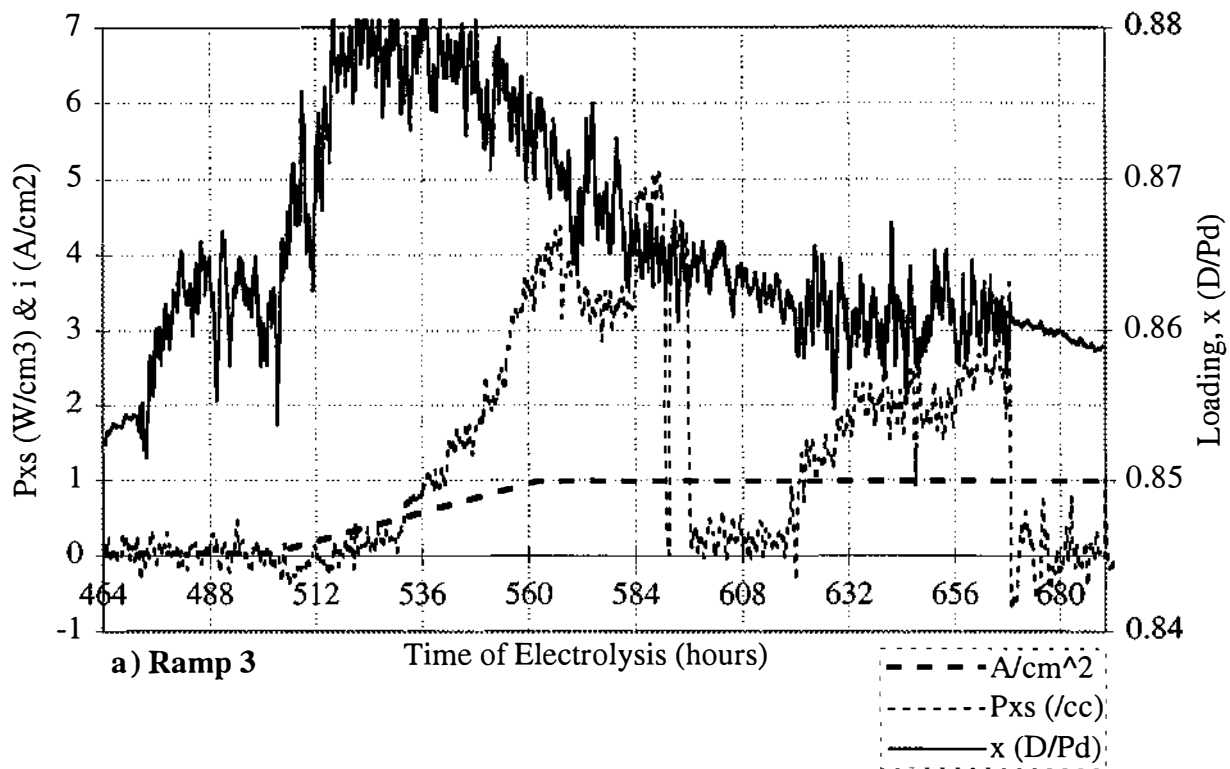
Figure 1 shows, initially, the normal response of a cell producing excess power:  $P_{xs}$  rising with increasing current density and loading above threshold values. After  $\sim 2$  days, however, the characteristic of  $P_{xs}$  at constant current density and (generally) decreasing loading, is unexpectedly dynamic. Furthermore excess power was observed in this cell at an unusually low maximum loading ( $D/Pd \approx 0.88$ ).

Examining closely the loading plotted in Figure 1 we see that this too is unexpectedly dynamic. For the first and last 24 hours shown, the amplitude of the variation in  $x$  is small, in the period around 608 h intermediate, but for the rest of the time the average loading shows a significant variation about the mean, with standard deviation  $\sim \pm 0.002$ . These periods of greater dynamism in  $x$  correlate with those for which  $P_{xs} > 0$ .

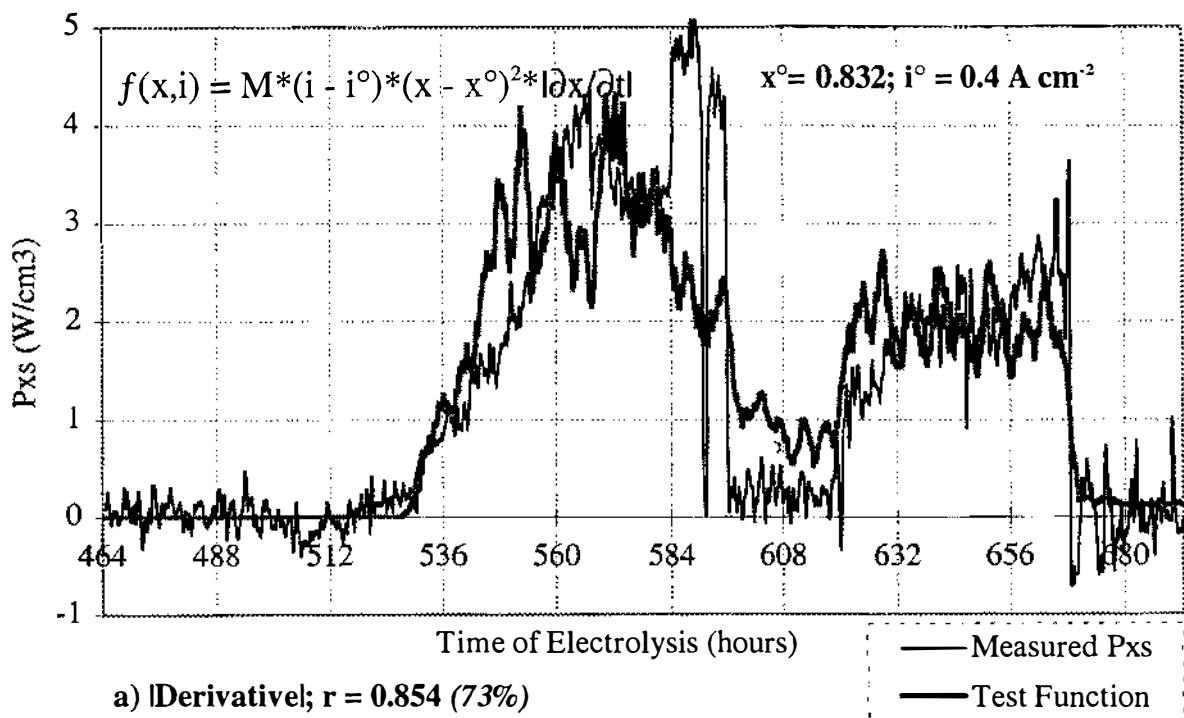
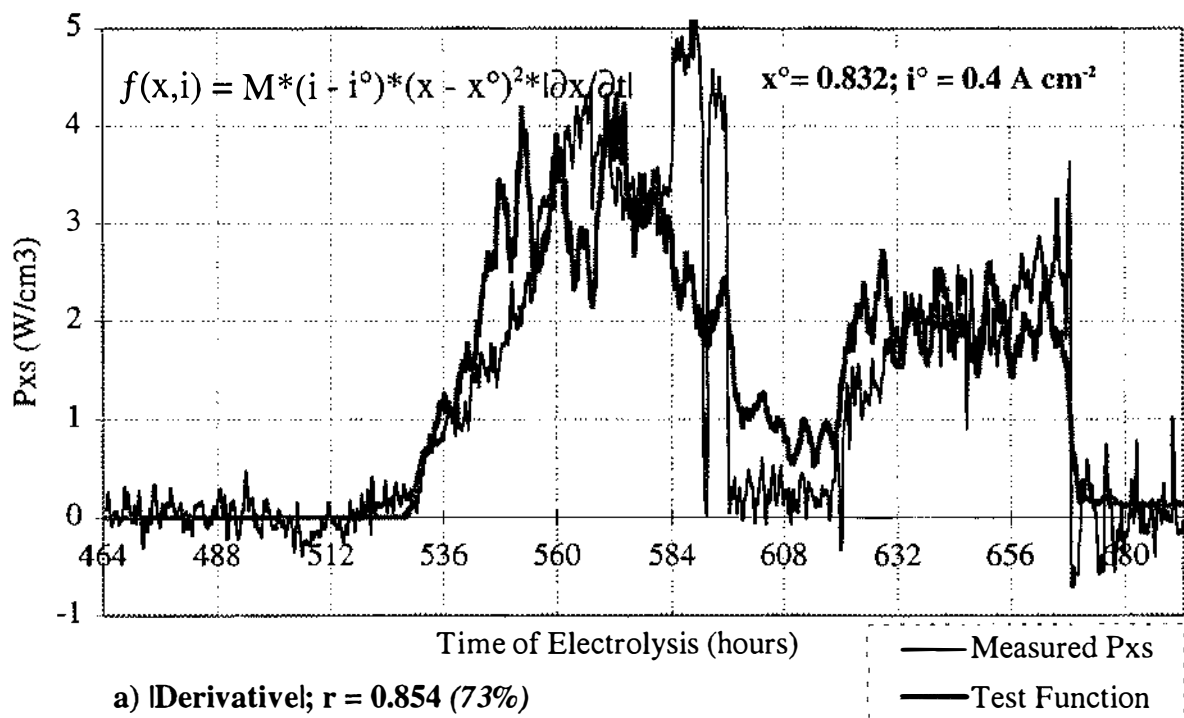
Figure 1b shows in detail the period of transition between low and modest excess power after 608 hours of electrolysis. While the frequency of the oscillation in loading does not change significantly, during the time of increasing  $P_{xs}$  (at  $t > 620$ h), the amplitude of this oscillation increases by a factor of 3 or more. When converted to a flux the rate of change of net loading could be accommodated by an adsorption and desorption current density, of  $\sim 0.1 - 1 \text{ mA cm}^{-2}$ .

Understanding that considerable approximation is involved, we will propose a simplified predictive function for  $P_{xs}$ , and test this against the time series data. Figure 2 shows the excess power data from Figure 1a compared to the test function,





**Figure 1 M4 Excess Power Current Density and Loading**



**Figure 2 M4 Excess Power and Fit Functions**

$$P_{xs.test} = M (i - i^{\circ}) (x - x^{\circ})^2 |\delta x / \delta t|$$

The term  $|\delta x / \delta t|$  assumes the importance of flux to be independent of sign, and gives no weight to a steady state flux. The proportionality constant,  $M$ , was determined to be  $2.33 \times 10^5$  in order to set the two functions  $P_{xs}(t)$  and  $P_{xs.test}(t)$  to equal energy. The threshold values,  $x^{\circ}$  and  $i^{\circ}$  were determined by maximizing the correlation between these two functions. For the two data sets shown in Figure 2, the correlation coefficient,  $r = 0.854$ , with  $x^{\circ} = 0.832$  and  $i^{\circ} = 0.4 \text{ A cm}^{-2}$

A value of 0.854 indicates that  $\sim 73\%$  of the excess power is related linearly to our test function. Other variables may be involved (the test function is not complete) or the coefficients may not be precisely right or the component variables strictly independent (the test function is not completely correct); this is nevertheless a remarkable degree of correlation when the approximations and implications involved in generating the test function are considered.

A factor not taken into account in the simple correlation function is the possibility of temporal displacement between the two data sets. If one imagines that the test function is a generating function and the measured excess power is the response (that our test function is causal) then one might expect  $P_{xs}(t)$  to be delayed with respect to  $P_{xs.test}(t)$ , and to have large amplitude (high frequency) features somewhat smoothed. Close inspection of Figure 2 reveals that this may indeed be the case; an analysis of the cross correlation function will be presented elsewhere, together with a more rigorous description of the treatment of the variable  $|\delta x / \delta t|$ .

Important questions are raised by the apparent success of our test function:

- i) how generally applicable is this function?
- ii) is the function predictive or responsive to other (possibly hidden) variables?
- iii) can the function be used to explain the appearance of excess power in some experiments and its non-appearance in others.
- iv) can the function variables be used to induce controllable excess power?
- v) what can this function teach us about the phenomenon under test?

On the question of general applicability, we are limited in our choice of comparative experiments. It would be desirable to select reference experiments having the same cathode geometry and dimension as the M4 cathode. Very few of our experiments have been performed with 1 mm wires, and none, previously, with the "lasso" geometry employed in M4. In practice, we are more constrained in our choice by the need for high data quality in resistance measurements, so that random measurement errors are not introduced into the values of  $\delta x / \delta t$ . Simply because the signal-to-noise ratio for 1 mm wires is better than the 3 or 4 mm diameter wires more typically (and successfully) employed, we are reduced in our selection of comparative experiments to one only: C1.

Experiment C1 has been described previously.<sup>4</sup> Figure 3a presents the loading and current density data for the first current ramp of C1. As for M4, the loading inferred from the measured resistance initially shows little perturbation, first decreasing with time at low current density, then increasing with the current ramp. Some time after initiation of the current ramp, an oscillation appears in  $x$ , which builds in amplitude. Figure 3b

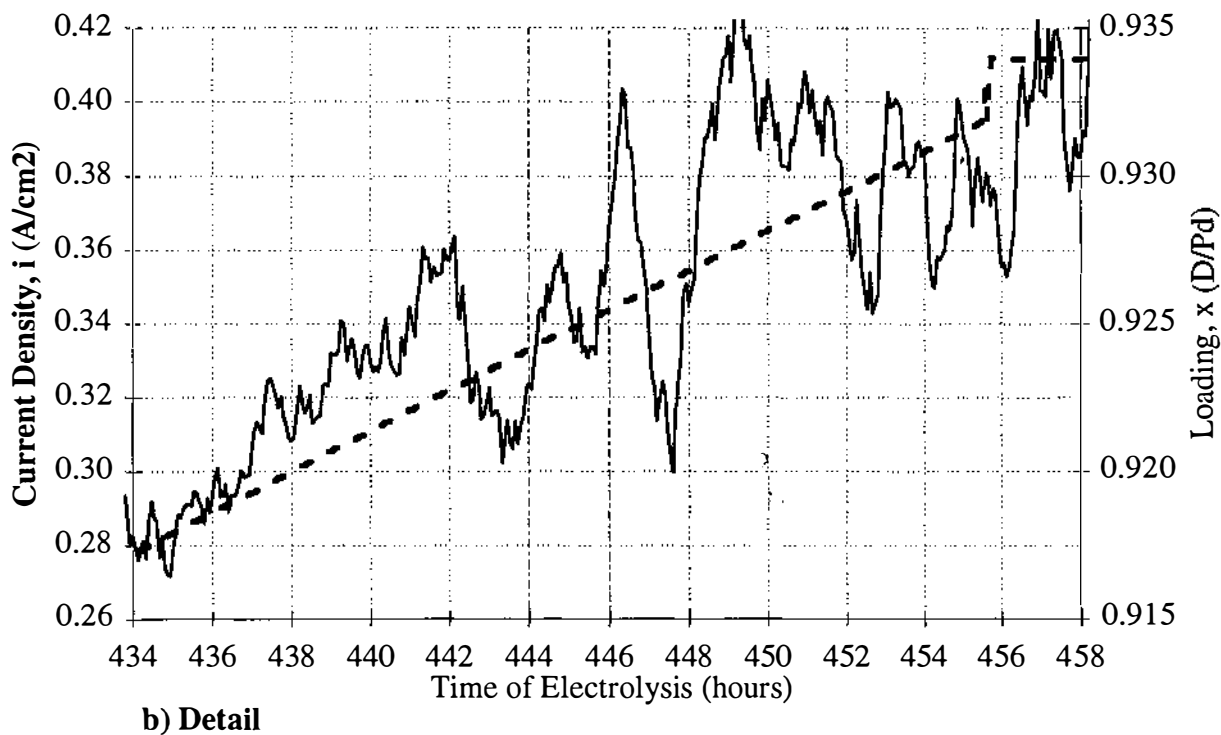
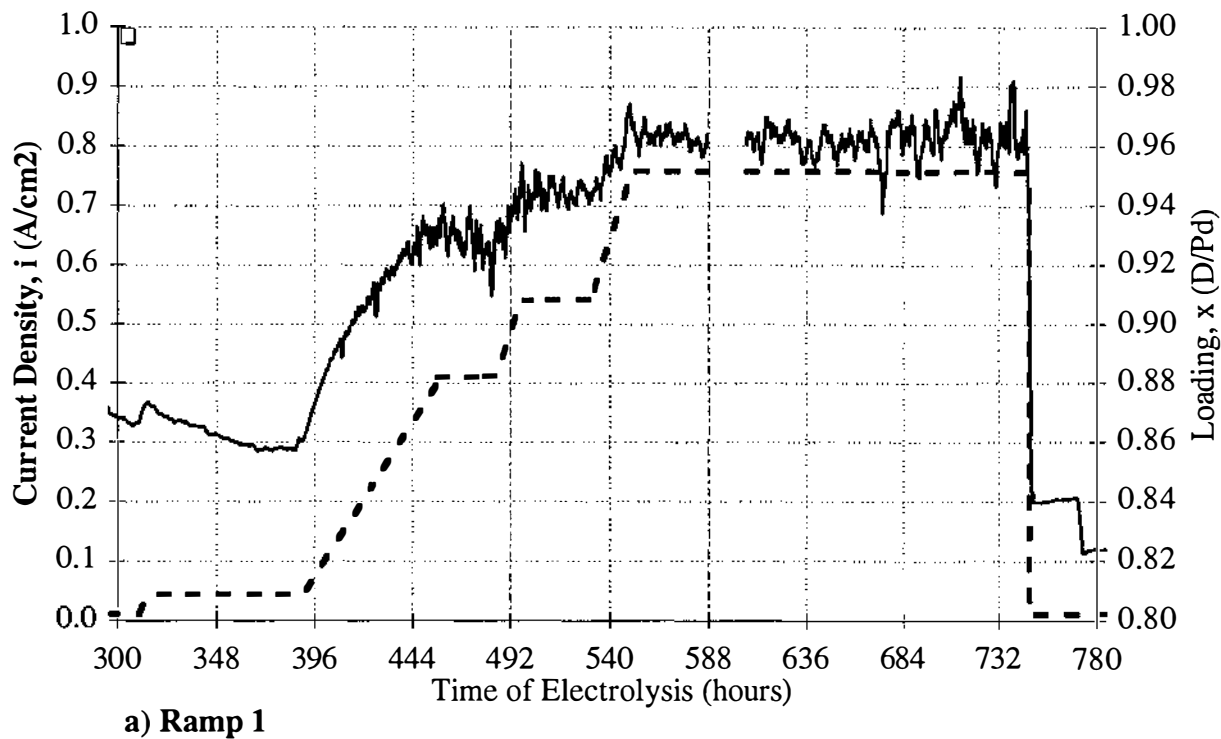
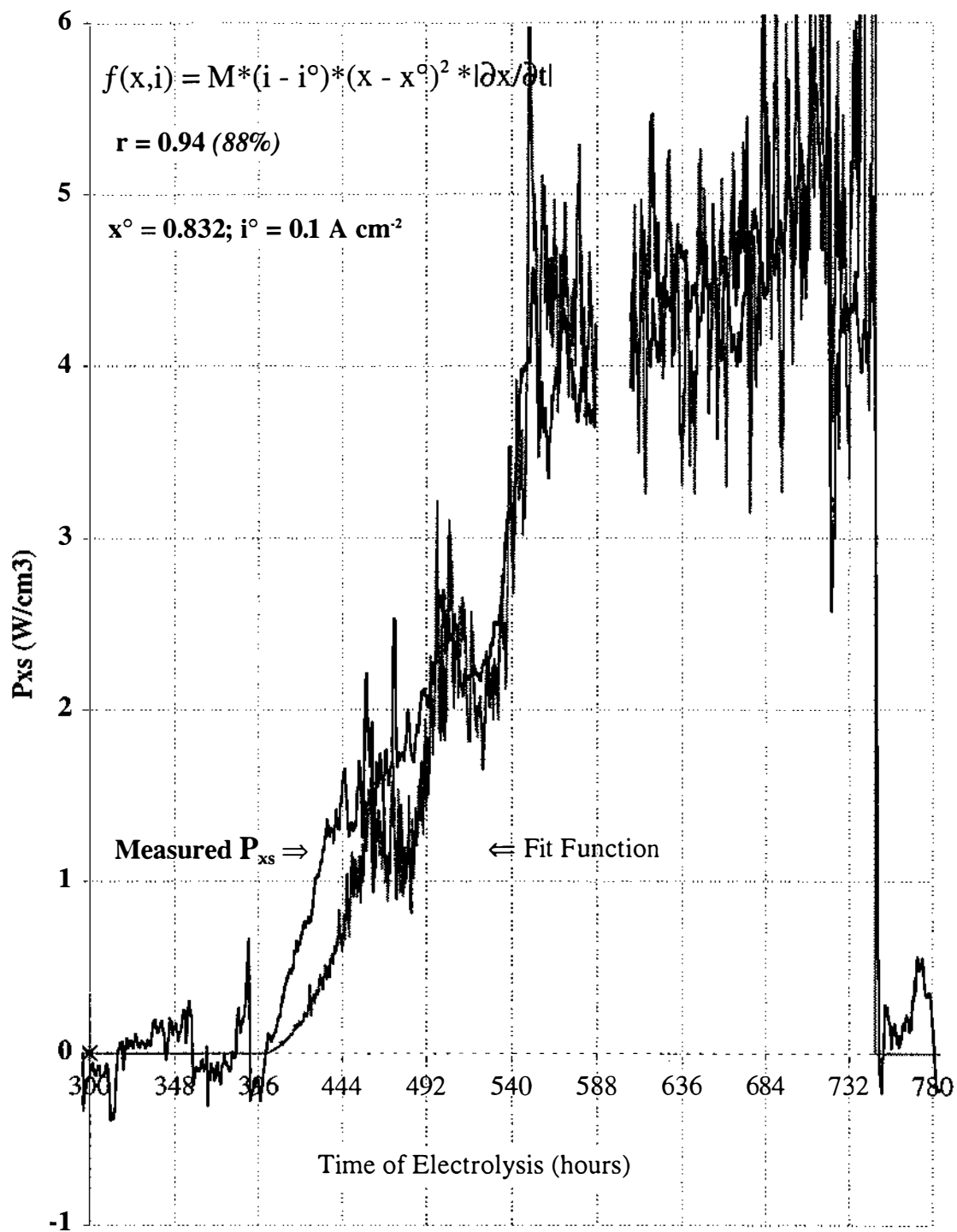


Figure 3 C1 Current Density (A/cm<sup>2</sup>) & Loading (D/Pd)



**Figure 4 C1 Excess Power and Fit Function**

shows a 24 hour detail in the vicinity of 444 h. While generally increasing with increasing current density, before 442 h the loading shows small fluctuation. After ~ 442h the loading exhibits a superimposed somewhat sinusoidal oscillation of period ~2h, as seen for M4. Perhaps significantly, the rate of increase in loading with increasing current density decreases at this point, suggesting the initiation of a transient de-loading (desorption) process, superimposed on the steady state loading.

Figure 4 shows the excess power measured during C1; ramp 1, compared to the test function employed previously for the M4 data (Figure 2). In this case, the value of  $x^\circ$  was chosen to be the same as previously used for the M4 data (0.832); this value is also consistent with the number found by direct regression of  $P_{xs}$  vs.  $(x - x^\circ)^2$ . The maximum correlation is, however, found with a significantly lower current density threshold for C1 (0.1 A cm<sup>-2</sup>) than for M4 (0.4 A cm<sup>-2</sup>). For the two data sets shown in Figure 4, the correlation coefficient  $r = 0.94$ , with  $x^\circ = 0.832$  and  $i^\circ = 0.100$  A cm<sup>-2</sup>. This correlation suggests that 88% of the function  $P_{xs,test}(t)$  is reflected linearly in  $P_{xs}(t)$ .

## 7. Discussions and Conclusions

We have demonstrated a mathematical correlation between excess power and the product of three variables: the excess current density,  $(i - i^\circ)$ ; the excess loading, squared,  $(x - x^\circ)^2$ ; the rate of change of the Pd cathode resistance. While we have demonstrated this correlation only for two experiments, M4 and C1, we have no reason to suppose that this correlation is not general. In one of two possible interpretations we have associated the rate of change of resistance with the rate of change of cathode deuterium loading,  $\delta x / \delta t$ , at constant temperature.

In terms of the "excess" parameters, there is little difference in the maximum value,  $(i_{max} - i^\circ)$  between experiments M4 and C1. Although the threshold value for M4 is much higher, the current density obtained in experiment M4; ramp 3, also was higher, and the exponent of 1 makes this not a strong variable. A significant difference does exist in the maximum "excess loading" variable,  $(x_{max} - x^\circ)^2$ . Due to the higher loading attained, this variable for C1 exceeded that for M4 by nearly an order of magnitude.

This latter observation is exceedingly important. Assuming that we can, as seems reasonable, use our test function as a predictor or, at least diagnostic for  $P_{xs}$ , then we need to pay close attention to the variables which give rise to large increases in the magnitude of the test function. An order of magnitude is the difference between indiscernible levels of excess power (~ 50 mW), and an interesting effect. This focuses attention on the need to obtain loadings as high as possible, above the threshold, as the parabolic dependence has greater power in amplifying the effect.

In the M4 and C1 experiments, the observed excess power densities and excess current densities were very similar. In order to achieve this result in terms of our multiplicative test function, the order of magnitude lower excess loading in M4 must be compensated for by a similar increase in the flux variable,  $\delta x / \delta t$ . In terms of hypothesis (ii), therefore, it is only the adventitious presence of a large deuteron flux that has promoted the excess power in experiment M4 from an insubstantial level at the low maximum loading achieved, to significant levels.

It is important to recognize that in neither experiment, M4 or C1, was an attempt made to maximize  $\delta x / \delta t$ . Quite the contrary. Both experiments were operated at constant (or slowly changing) currents, temperatures, and gas pressures; the three variables most

likely to influence loading. What fluctuation in loading did occur, and in both cases it was significant and varied, occurred apparently spontaneously; we observed the effect, we did not control it.

Nevertheless, if our hypothesis is correct, it may prove advantageous to understand the role of deuteron flux, and the means by which this can be stimulated without net de-loading, as a means to understand the phenomenon of excess power generation.

To this point we have treated the data statistically, and empirically. The functionality of our excess power test function can, however, be discussed in terms of at least one of the theories proposed for "cold fusion" phenomena. Hagelstein<sup>13</sup> has proposed that a phonon laser operates to initiate solid state neutron transfer, and to couple the nuclear energy produced, to the lattice, as heat. The details of this model are presented in Reference 13, and in preceding papers cited therein. This model makes two predictions that are relevant in the discussion:

- i) No excess power will be observed at deuterium loadings below that at which the partial molar enthalpy change for desorption becomes exothermic.
- ii) The rate of excess heat release (excess power) will increase with the desorption flux.

The first (threshold) criterion establishes the point at which a phonon laser may begin to operate. In an accompanying paper<sup>14</sup> we have attempted to define the position of this loading threshold based on literature data for the Pd-D system. By extrapolating the literature data at lower loadings, we estimate that the threshold value for the exothermic desorption of deuterium from palladium to be  $0.83 \leq x^0 \leq 0.85$ . A value closely in accord with our observed value of  $x^0 \approx 0.832$ .

The second (flux) criterion is associated with the rate of phonon excitation. This flux can be measured as the rate of change of the average loading; in the Hagelstein model only the desorption flux plays a role. In our empirical function we have employed  $|\delta x / \delta t|$ . For a symmetric loading/de-loading cycle (*i.e.* no dc term) the average value of  $|\delta x / \delta t|$  is simply twice the average value of the desorption component alone, so that our observed correlation would be unchanged, and consistent with the Hagelstein model prediction.

A great deal more needs to be said about our results, the correlation analysis, and the relationship of these to the Hagelstein model predictions. We need to consider the time-scales of the various processes, the relationship of the observed transient net flux to the atomic scale process of a heterogeneous surface, and any possible role that a steady state flux may play. A related topic of importance is the relationship between the endothermic/exothermic transition surveyed by Crouch-Baker<sup>14</sup> to the phonon laser threshold predicted by Hagelstein<sup>13</sup>, particularly as these affect the onset of thermal positive feedback, observed calorimetrically by Pons and Fleischmann.<sup>15</sup>

Despite our incomplete understanding, we are encouraged to see, perhaps for the first time in this field, the suggestion of a synthesis and consensus of theory and experiment.

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