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OVERVIEW AND STATUS OF THE EPRI PROGRAM ON DEUTERATED METALS

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ABSTRACT

Some five years have elapsed since the first announcement by Fleischmann, Pons and Hawkins (1989) of the observation of excess heat from palladium heavily loaded with deuterium by electrochemical polarization in heavy water in which lithium deuteroxide served as electrolyte. The EPRI program began in April, 1989, and has continued to the present time attempting to replicate the claimed excess heat and determine its source. Under conditions difficult to achieve, some 15 separate experiments have successfully reached that goal out of some 30 major attempts. The conditions found necessary for an observation of excess heat were three in number: 1) atomic loading ratio (D/Pd) > ~0.9; 2) Initiation time of 8 to 23 days; 3) current density >0.1 amperes per cm2 of cathode area. No definitive source for the excess heat has been yet robustly determined, but measurable helium-4 has been observed in the cell vapor space in a few cases. The major evidence that the heat may be from nuclear reactions is its magnitude - some 10 to 100 times larger than any known chemical reaction. The objective of the continuing effort is focussed upon identifying in a roughly quantitative manner, the source of the excess heat.

INTRODUCTION

The subject of cold fusion has aroused considerable controversy since its announcement some five years ago. This controversy was to be expected in any proposed new potential energy source that would threaten the position of research on existing and other potential energy sources. This atmosphere of controversy plus the natural tendency of inventors to keep secrets has slowed the resolution of the scientific issues. The EPRI program had two major objectives at the outset: 1) to obtain the largest possible amount of excess heat; and 2) to observe the neutrons and tritium everyone expected from the D + D fusion reaction. Since that time the focus has not shifted on the excess heat issue but the search for neutrons and tritium has shifted to a search for helium-4 and possible isotopic abundance shifts among stable nuclei within the cathode. Several credible episodes of neutrons and tritium were seen in the EPRI-sponsored work and by others (Storms, 1991) but at levels far too low to be the primary heat-producing reactions, and in most instances, from experiments not designed to measure excess heat.

THE SEARCH FOR EXCESS HEAT

Over 70% of the effort by EPRI has been directed to understanding the process that produces excess heat. This work has been extensively reported by McKubre and coworkers at SRI International (1994 and 1993). The initial strategy was to get so much heat as to be unequivocal and to exploit its use even before the source of heat was known. This was logical because the excess heat already appeared to be one or two orders of magnitude greater than any known chemical process within so small a mass of material (palladium). Therefore it was presumed to be a nuclear reaction providing the heat. Subsequent research has shown that it must also be an entirely different reaction pathway than the one widely explored in plasma fusion research because a requisite level of neutrons and tritium are obviously not

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present. However, the small but definite levels of neutrons and tritium credibly observed by a few investigators (Wolf et. al. (1991), Will, Cedzynska, and Linton (1993), Tuggle, Claytor and Taylor (1994) and Takahashi et. al. (1992)) are indications that some unexplained phenomena are occurring in the palladium metal lattice - at levels some 40 or more orders of magnitude larger than existing theory predicts - even though 6 to 12 orders of magnitude too small to explain the excess heat! Furthermore, the ratio of neutrons to tritium is 6 or more orders of magnitude lower than observed for the D+D reaction. This evidence is an indication that a nuclear process is indeed possible in highly deuterated palladium and that it is not the same as the ones widely studied in fusion research up to now. If enough excess heat can be generated, even subtleties such as isotopic abundance shifts of stable nuclides or helium-4 production will eventually be at levels large enough to measure readily and unequivocally.

Reproducibility is the issue continually raised by both critics and active researchers in the search for excess heat episodes. Clearly the conditions thus far observed to be necessary for achieving a positive result are not easily attained. Most deuterium charging of palladium by electrochemical means reaches a D/Pd ratio of 0.7 but not higher without careful manipulation of electrochemical and cathode metallurgical conditions. It is thus understandable that not everyone who has attempted the experiment, especially those operating within a limited time horizon of a few months, gets the effect. Since hydrogen charging of, and diffusion within, palladium has been intensively studied for over 100 years without discovery of the anomalaous heat effect, the conditions for it must be very rarely achieved. This is indeed the experience of the few research groups who have claimed success - that is, a large fraction of the experiments are not successful. One significant reason for such a situation is the issue of achieving the apparently required D/Pd loading ratio. Pons (1992) has remarked that a batch to batch variability has been observed among palladium used for cathodes. In one batch, a significant fraction of cathodes give the effect, whereas in another batch, none of the cathodes show excess heat. This effect is reminiscent of the heat to heat variability in stainless steels in their susceptibility to stress corrosion cracking, usually due to variations in the previous heat treatment and carbon content..

Figure 1 gives the excess heat results as a function of the resistance ratio of the palladium cathode, the means by which the D/Pd ratio is almost continually monitored (McKubre et al. (1994). Note that above a maximum achieved loading of 0.95 every cell gave excess heat whereas below a D/Pd ratio of 0.9 no cell gave the effect. In the gap between these peak loading ratios about half the cells showed the anomalous heat effect.

Table 1 gives a summary of results of some 26 major attempts by McKubre and coworkers (1994) to replicate the excess heat phenomenon claimed by Pons, Fleischmann, and Hawkins. (1989). As an illustrative example of one of the cells in Table 1, Figure 2 gives the fraction of excess power in cell P-15 over a ~100 hour period. The fraction is calculated two ways - 1) relative to the total input power which includes a compensating joule heater to maintain isothermal conditions during changing electrochemical conditions and 2) relative to the electrochemically generated power alone. Relative to the latter the excess maintains about a 10%level with a peak at ~25%. Figure 3 is a corresponding curve of the cathode resistance ratio for P-15 during the same period as Figure 2 relative to pure, unloaded palladium. A resistance ratio of 1.58, the lower plateau on the curve corresponds to a D/Pd ratio of 0.97. Similar data for cell P-16 is also shown in Figure 3. Note that P-16 achieved a higher resistance ratio of 1.6 corresponding to a D/Pd ratio of 0.94. Table 1 indicates P-16 also showed a lower excess heat effect than P-15.

Another issue frequently raised is the one of light versus heavy water. Figure 4 gives a side by side comparison between cells P-13 (light water) and P-14 (heavy water) over a 200 hour period (McKubre et al. (1994)) The two cells operated in series electrically in the same water bath and were monitored by the same instruments on a time shared basis. While the loading ratios for H and D of the two cathodes were not precisely the same, there was a clear difference in excess heat favoring the heavy water cell. Within the limits of error, cell P-13 registered near zero excess heat. It is an apparently valid criticism that an inadequate number of so-called "blanks" with light water have been run. The investigators' response has been that an even better "blank " is a heavy water cell that produces no effect. There is some evidence from work elsewhere that light water is also effective in producing excess heat in cells with nickel cathodes and alkali carbonate electrolytes. The real reason for the small number of "blanks" has been the restrictions from the limited level of available experimental resources giving pressure for more "positive" results.

Not shown here is one case in which an operating heavy water cell exhibiting excess heat is injected with enough light water to raise to 10% the fraction of light water in the electrolyte. In about 100 hours the excess heat died away to near zero.

SEARCH FOR NUCLEAR REACTIONS IN Pd

About 30% of the total EPRI effort has been focussed upon the search for nuclear products such as neutrons and tritium. No tritium above the background level present in the heavy water electrolyte was seen in any of the excess heat experiments shown in Table 1. A low level neutron capability was not in place for those experiments.

Wolf, et. al.(1991) reported low level neutron episodes in deuterium loaded palladium. However, in no case has the level been more than several times the background level from cosmic rays. Nonetheless, this is still some 40 or more orders of magnitude greater than expected from calculations for the presumed normal deuteron spacings in the palladium lattice.

More recently some nuclear reactions of a more definitive nature have been observed in deuterated palladium but their open publication awaits a replication of the results. (Wolf, (1992)). Here, reproducibility has been particularly elusive, perhaps even more so than with the observation of excess heat. Batch to batch variations in the palladium cathode material in this case may not be the source of the problem since most experiments have used samples from only one batch.

CURRENT STATUS

Current batches of palladium appear to be difficult to load to the previously determined required level for an excess heat episode. Therefore, the current effort searching for increased excess heat is focussed upon finding what uncontrolled variable is preventing achievement of high loading. Impurity content of the most successful palladium batch appears to be slightly higher than the less successful batches. Most of this impurity

content is the remnant of materials added during the final pouring of the ingot to remove oxyge. Calcium boride being the primary constituent with a significant silicon level as well. One hypothesis is that oxygen content remaining in the palladium after production is a significant factor preventing full loading. The mechanism suggested is analagous to the one for avoiding oxygen in copper used in reducing environments - to avoid porosity created when hydrogen reacts with such oxygen to produce water. Usually oxygen is not measured among the impurities so there is as yet no definitive proof of this hypothesis.

Efforts are continuing to attempt replication of the nuclear reactions previously seen apparently connected with low level neutron emission (Wolf (1993)). If and when successful, this effort would provide unmistakable and unambiguous evidence of nuclear reactions in palladium in electrochemical cells at high deuterium loadings, whether connected with heat production or not.

CONCLUSIONS

Evidence for low level nuclear reactions in heavily deuterated palladium is accumulating from many different laboratories but not nearly enough to explain the few watts of excess heat assuming the D + D reaction is occurring. Either some other reaction is the heat producer or the D + D reaction has been entirely changed from its known characteristics so that the reaction pathway favors He-4 and does so without the 23.8 Mev gamma ray being emitted.

Because the batch to batch variation in both the production of excess heat and tritium is so prominent, it is not unlikely that some of the major impurities are the source of the heat and helium-4 observed. The most likely suspect in this category is boron since the d, alpha reaction on B-10 produces three He-4 atoms with a positive energy release of 17.8 Mev. The most likely impurity for tritium production is beryllium which exhibits a D,T reaction with a positive energy release of 4.6 Mev. Beryllium has been found at the PPM level in a few palladium cathodes used in EPRI's program.

The search for the source of the excess heat will continue to dominate EPRI's interest because one of the power industry's major businesses is the conversion of heat to electricity. If it turns out that a nuclear reaction is proved to be the source of the excess heat, then it would remain to find the laws for scale up to practical levels. However, that scaleup effort could occur with the assurance of a very large supply of fuel far beyond any existing ones in current use.

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LIST OF FIGURES AND TABLES

Table 1. OVERVIEW OF CALORIMETRIC RESULTS FROM 26 ELECTROCHEMICAL CELLS

#	Electrode A (cm)	B (cm)	C metal	D	E	F (M)	G	H °C	I (psi)	J	Maximum Loading R/R°	D/Pd H/Pd	Calorimeter type	Duration Expt. (h)	Init. (h)	Maximum Power Input (W)	Excess (W)	%	Total Input (MJ)	Energy Excess (MJ)	%	K #
Differential:																						
P1a	5	0.7	AECL		LiOD	1.0		7	650	682	1.20	1.06	Bulk	696	369	3.35	1.75	52.2%	3.4	0.07	2.1%	5
P1b	5	0.7	EP		LiOD	1.0		7	650	682	?	?	Plate	696	299	3	0.2	51.0%	3.0	0.02	0.7%	2
Mass flow:																						
P2	4.5	0.4	JM	AR	LiOD	1.0		4	1000	495	1.65	0.94	Si-oil	1393	504	3.8	2	52.6%	50.2	1.07	2.1%	4
P3	4.5	0.4	JM	AR	LiOD	1.0		25	1000	265	1.70	0.92	Si-oil	1250					18.0			0
P7	4.5	0.3	E#1	AR	LiOD	1.0		8	1000	259	?	?	Si-oil	145					2.1			0
P4	5	0.3	E#1	AR	LiOD	0.1		30	100	509	1.80	0.88	Si-oil	1165					16.8			0
P5	5	0.3	E#1	AR	SO4	0.5		16	100	849	1.70	0.92	Si-oil	287					4.1			0
P6	5	0.3	E#1	AR	SO4	0.5	As	4	100	573	1.70	0.92	Si-oil	649					9.3			0
P8	3	0.3	E#1	AR	LiOD	0.1		35	100	637	1.65	0.94	Si-oil	186					2.7			0
P9	3	0.3	E#1	AR	LiOD	1.0		35	52	531	1.65	0.94	Si-oil	597					21.5			0
P10	4.5	0.4	JM	AR	LiOD	1.0		35	900	47	?	?	Si-oil	18					0.3			0
P11	4.5	0.4	JM	AR	LiOD	1.0		35	1025	1179	1.65	0.94	Si-oil	85					1.2			0
P12	3	0.3	E#1	4He	LiOD	1.0	Al	30	50	884	1.55	0.97	Si-oil	1631	316	10	0.97	9.7%	58.7	0.80	1.4%	4
P13	3	0.3	E#1	AR	LiOH	1.0	Al	30	50	884	1.10	0.97	Si-oil	815		15	0	0.0%	11.7	0.00	0.0%	0
P14	3	0.3	E#1	3He	LiOD	1.0	Al	30	50	884	1.60	0.95	Si-oil	692	184	10.5	0.5	4.8%	10.0	0.20	2.0%	2
P15	3	0.3	E#1	AR	LiOD	1.0	Al	30	40	884	1.58	0.96	***	1104	684	10	2.4	24.0%	39.7	0.55	1.4%	3
P16	3	0.3	E#1	3He	LiOD	1.0	Al	30	40	884	1.70	0.92	***	1104	948	10	0.4	4.0%	39.7	0.10	0.2%	4
P17	3	0.3	E#1	AR	LiOD	1.0	Si	30	40	389	1.29	1.04	water	1202	1040	10	0.2	2.0%	13.0	0.10	0.7%	2
P20	3	0.3	E#1	AR	LiOD	1.0	Al	35	40	707	1.55	0.97	water	954	650	12	0.28	2.3%	17.2	0.16	1.0%	3
P19	3	0.3	E#1	AR	LiOD	1.0	B	35	40	672	1.45	0.99	water	1287	261	0.25	0.85	340%	42.0	0.79	1.9%	5
P21	3	0.3	E#1		LiOD	1.0	B	30	40	707	1.60	0.95	water	764	390	10.5	0.6	5.7%	13.8	0.04	0.3%	2
P22	3	0.3	E#1		LiOD	1.0	B	30	40	707	1.30	1.03	water	1480	378	0.27	0.08	30%	21.3	0.27	1.3%	3
C1	30	0.1	E	AR	LiOD	1.0	Al	30	50	764	1.65	0.94	water	866	390	45	1.35	3.0%	49.1	1.12	2.3%	1
C2	100 µm		JM	AR	LiOD	1.0	Al	30	50	120	1.60	0.95	water	356	190	35	3	8.6%	14.4	0.56	3.9%	1
L1	3	0.3	E#3	AR	LiOD	1.0	Al	30	20	1088	1.99	0.76	water	1600		50			95.0			0
L2	3	0.3	E#3	AR	LiOD	1.0	Al	30	20	1088	1.99	0.76	water	900		30			50.0			0

Electrode characteristics:

A. Length. B. Diameter. C. Metal Source. AECL = Atomic Energy of Canada Limited;
EP = electroplate. JM = Johnson Matthey; E#1 = Engelhard lot #1. E = Engelhard; E#3 = Engelhard lot #3
D = surface preparation: AR = aqua regia rinse; 4He = ^{4}He implantation; 3He = ^{3}He implantation

Electrolyte characteristics:

E. Electrolyte type (SO4 = Li_2SO_4). F. Electrolyte concentration. G. Additives (at 200 ppm)

Physical characteristics:

H. Bath Temperature. I. Gas pressure (D_2 or H_2). J. Maximum current density
Maximum loading determined from resistance ratio (R/R°)
Calorimetry fluid Si-oil = silicone oil. Water = H_2O

Excess power observations:

K. Number of instances of excess power observed in that experiment

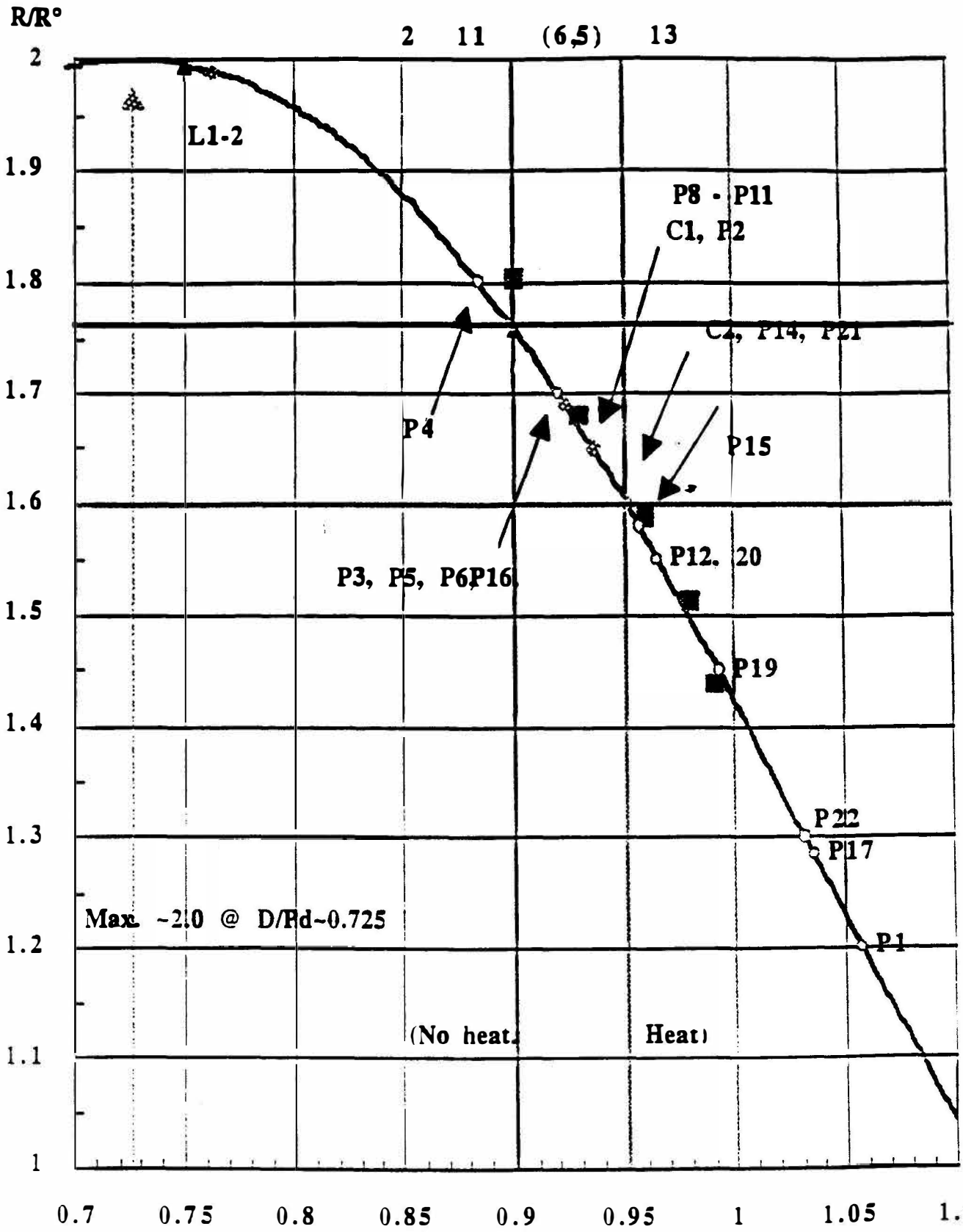

Figure 1 Maximum loading, D/Pd, attained in experiment: determined by R/R°.

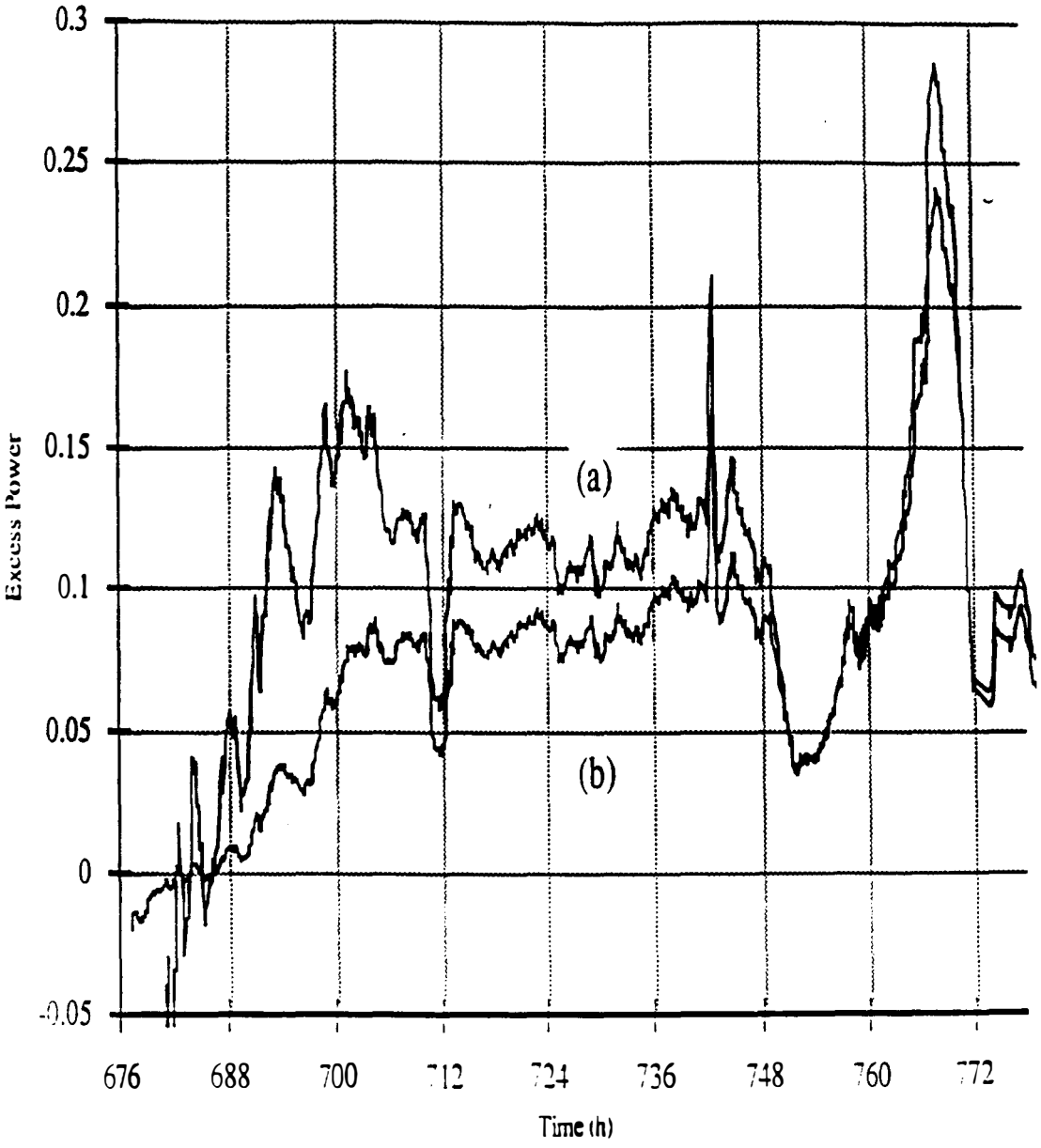

Figure 2
Variation of excess power for P15 with time (since start of experiment), expressed (a) as a fraction of the electrochemical power, and (b) as a fraction of the total input power

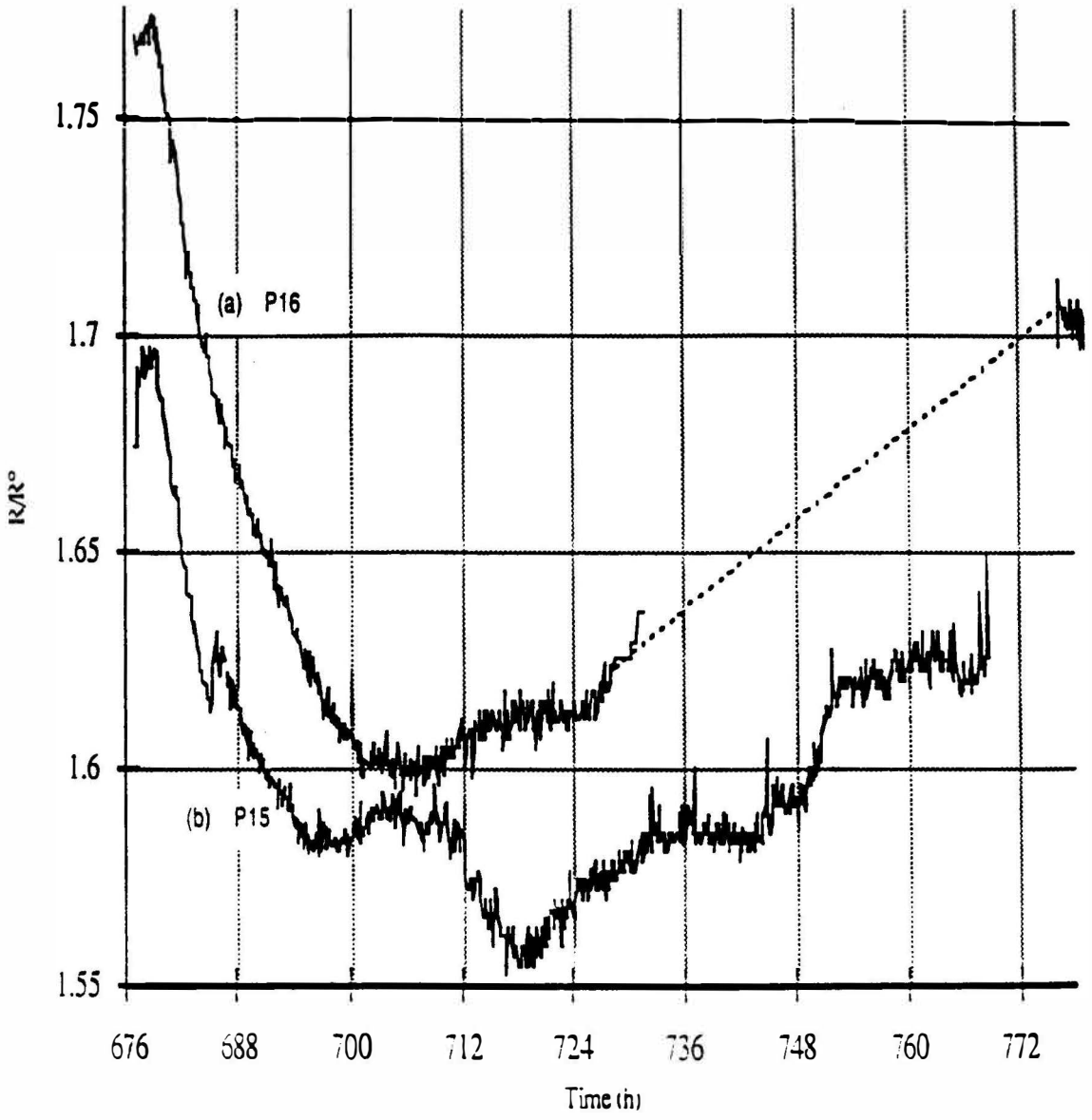

Figure 3
Resistance ratio variations for (a) P16 and (b) P15 with time (since start of experiment)

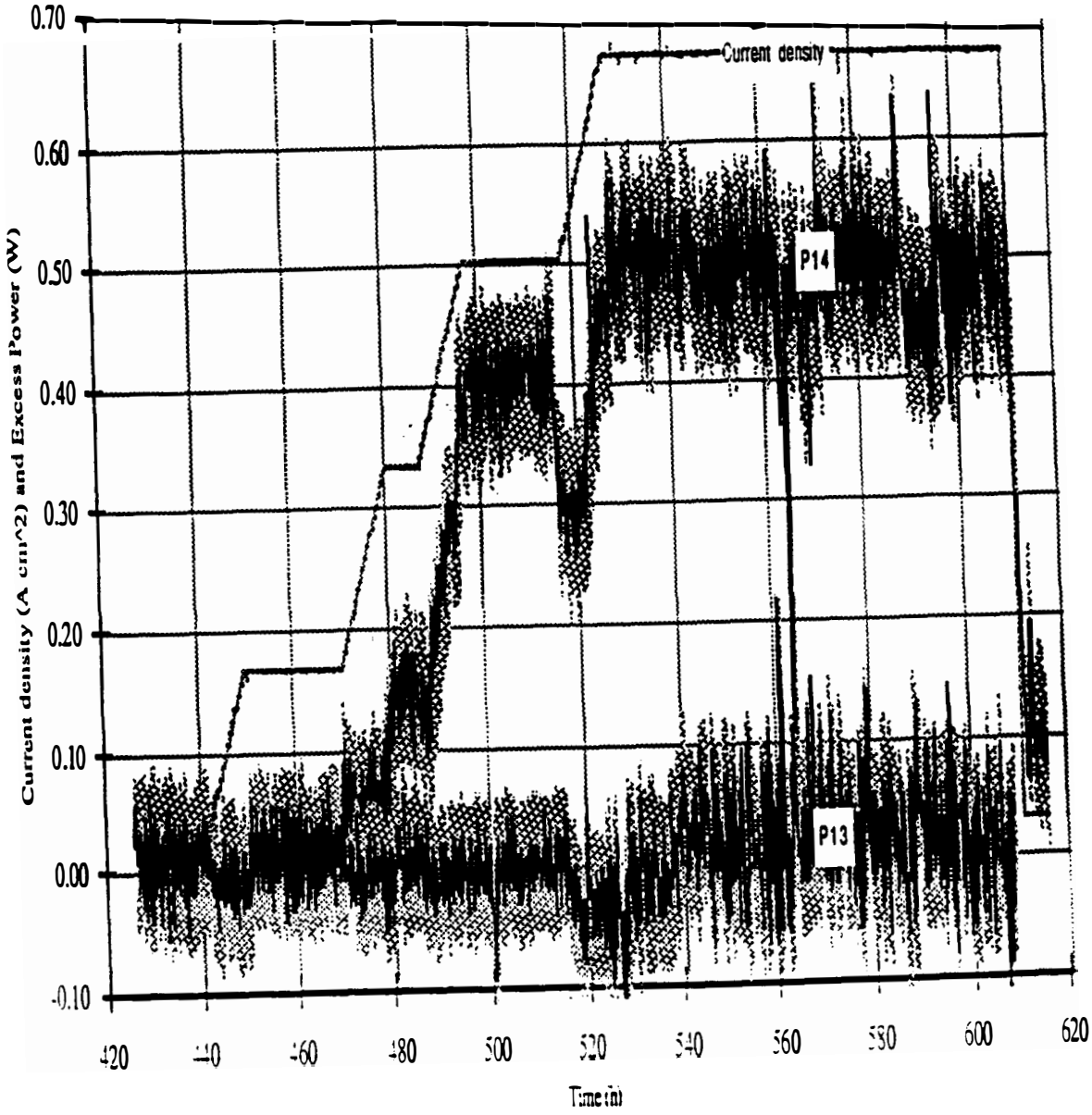

Figure 4
Variation of current density (A cm^{-2}) for P13 and P14, excess power (W) for P14, and excess power (W) for P13 with time (since start of P13). For each excess power curve (heavy line), the associated uncertainty span (hatched line) is superimposed