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# PROGRESS IN THIN-FILM LENR RESEARCH AT THE UNIVERSITY OF ILLINOIS

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## ABSTRACT

The research described here includes work on fabrication techniques for reproducible thin-film electrodes. Runs with these electrodes in a newly fabricated high sensitivity calorimetry bank is shown to provide added support for earlier excess heat production observed with ultra-high proton loadings in thin film electrodes. In addition, new in-situ radiation emission studies have discovered MeV alpha-proton emission, supporting earlier reaction product evidence of the nuclear origin of the excess heat. Recent experiments and lattice simulation studies have provided added insight into highly loaded thin film phenomena, including possible H- effects associated with anomalous resistivity effects at ultra-high loadings.

Research is continuing to study optimization of materials for maximum excess heat, lifetime, and minimum costs. Concepts have been devised to scale the present single electrode experiments up to specific powers of ~1W/cc cell-vol., using replication of electrodes in a closely packed multi-electrode configuration. The ultimate objective is to achieve 100-W to 20-kW units for distributed power network applications.

## 1. INTRODUCTION

In this presentation we briefly discuss the following:

- Comments about original bead packed-bed electrolysis and measurement of nuclear reaction products;
- Subsequent thin-film plate-type electrolysis used to study excess heat production and also MeV alpha-proton particle plus X-ray emission;
- New test cell calorimetric bank and thin-film plate heat studies;
- Supporting studies of anomalous resistivity effect in thin films at high loading;
- Comments about possible scale-up to higher power units.

Cold fusion research at the University of Illinois has largely concentrated on light water nuclear reactions, termed Low Energy Nuclear Reactions, or LENRs. Thus in contrast to D-D type reactions where $He^4$ is a major product, LENRs result in a wide array of reaction products ranging from light to very heavy masses.

As outlined in previous ICCF presentations [1-3], the key "signatures" of these LENR reaction products are:

- Large reaction rates for key elements;
- Excess heat rates of order of 2 – 10 W/cc of cell volume;
- Peak mass zones of high yields separated by low yield "gaps";
- Non-natural isotope ratios for many products;
- Low-energy (~20 keV) X-ray or beta radiation emission observed.

Research on thin-film electrode based electrolysis has continued to study film fabrication techniques needed to obtain reproducibility of results, parametrics of excess heat production, energetic charged particle and X-ray production, and anomalous resistivity effects vs. loading. The use of thin-films has remained the hallmark of this

research. This focus dates back to original studies of multiple-layers thin-film research reported in ICFF-4 [4]. The guidance for design and material's selection for the films remain the original swimming electron layer (SEL) theory [5].

## 2. THIN FILM FABRICATION AND QUALITY CONTROL

The conceptual design for the thin-film plate-type electrodes used in the studies described here is illustrated in fig. 1.

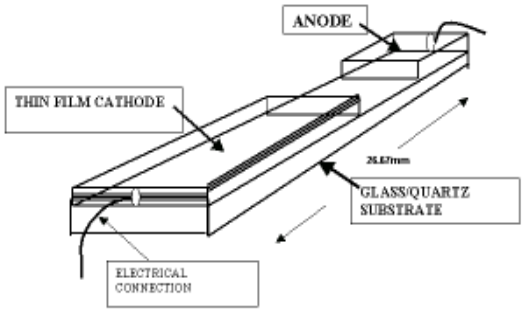


Fig 1. Conceptual design of plate-type electrode (in some designs the anode is a separated Pt. wire or plate rather than being sputtered on the substrate as shown here. Alumina has been selected as a "reference" substrate.

Reproducible results using thin-film electrode require careful control of the fabrication steps. The basic challenge comes from the large stresses created during electrolytic loading, which, in the extreme case can destroy the films. As discussed in ref. [6], fabrication research has studied parameter space in each step combined with a detailed analysis after each step to quantify results and identify optimum conditions.

Excellent films have been achieved as demonstrated by the X-ray pattern for a Ni-coated Pd film on $Al_2O_3$. The successful formation of the Ni-Pd film is confirmed by lines for the Pd [1 1 1] at ~ 40.178, Ni[1 1 1] at ~ 44.582, Pd[2 0 0] at ~ 46.733, and Pd[2 2 0] at ~ 68.241. Consequently, the Ni/Pd film on alumina has been selected as the reference electrode configuration for this work. Fabrication is done by first smoothing and cleaning the alumina substrate, pre-annealing it, pre-coating the substrate surface and then sputtering on the film, followed by controlled vacuum annealing.

## 3. NEW LENR CALORIMETRY BANK

A new calorimetry bank was recently constructed in the LENR lab to provide a means of rapidly testing series of electrodes using reasonably high sensitivity calorimetry with good accuracy. This bank consists of:

- Four isoperibolic calorimeters based on a high sensitivity (~4 degree/W) design for rapid testing of parallel multiple;
- Open cell electrolysis, each cell stirred magnetically, all coated in a constant temperature water bath;
- Two thermistors per cell (~0.02°C precision);
- Accuracy of ±20mW to ±40mW (low to high power);
- Dual calibration using resistor heating and reference electrode electrolysis;
- HP34970A acquisition data system.

The capability of this calorimetric bank is summarized in Table I. Most of the electrodes presently studied have heat inputs of order of 0.5 W of more and excess powers from 100 mW and above. Thus the accuracy of ±20mW at low powers to ±40 mW at high powers is considered quite adequate for this work. Further details about this bank and the use are given in ref. [7].

**Table I: Bank Characteristics**

| | |
|---|---|
| Thermistors | Accuracy - 0.1°C<br>Precision - 0.02°C (~5mW) |
| Bath Temp. | 0.05°C (~13 mW) |
| Thermistor ΔT | Less than 0.02°C (~5 mW) |
| Δ T ambient | ~0.02°C/°C (max. 20 mW) |
| Recombination | 3% |
| Electrolyte ΔV | ~3% (~5 ml volume correction) |
| Calorimeters | Error Analysis ~ ±30 mW<br>Exp.fit ~ ±20mW (to ±40 mW at higher power) |

## 4. RECENT ELECTRODE STUDIES USING NEW TEST BANK

The test bank described above has been used to evaluate a variety of new electrode designs. As an example, a trace of an excess heat study using the "reference" Ni/Pd electrode described earlier is shown in fig. 2.

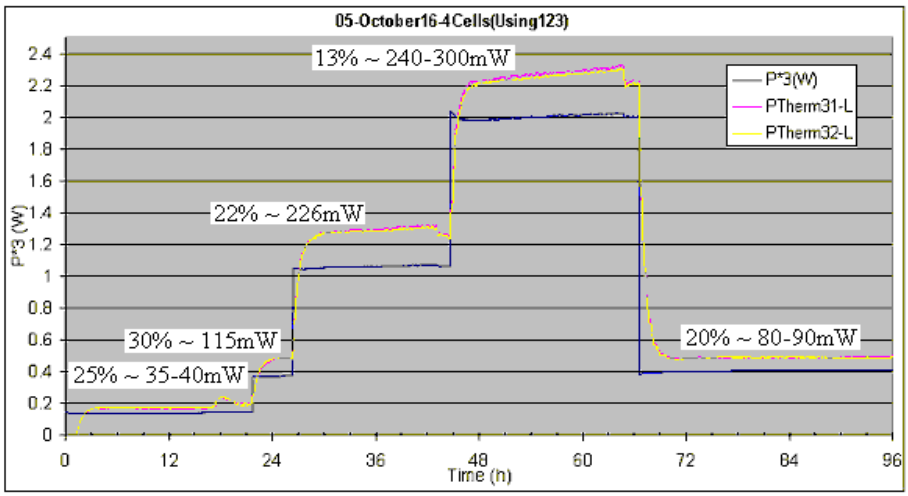


Fig. 2. Typical run with the reference Ni/Pd alumina electrode.

This electrode exhibited excess heat ranging from 13 to 30%, dependent on the input power. Details about the analysis of the data and evaluation of the excess heat values are given in ref. [7]. In particular, at higher input powers the absolute excess power is well about the uncertainty limits of the calorimetry, including allowance for a variety of errors ranging from temperature gradients to off-gas recombination. The electrode was examined after the 15-day run and found to exhibit some surface deterioration but no serious physical damage. Both larger and smaller excess heat production has been obtained with other film material combinations and designs. Evaluation of this data is still in process with the objective of simultaneously optimizing the percentage excess heat and electrode lifetime. The ease of fabrication and costs are also considerations in the electrode design selection.

## 5. CHARGED-PARTICLE AND X-RAY EMISSION STUDIES

In situ studies of charged-particle emission during electrolysis have been undertaken to supplement the earlier reaction product measurements. This work used small CR-39 sheets and TLD chips attached to the film surface to record charge particles and also x-ray production. The CR-39 tracks were calibrated using MeV apha and proton bombardment with high-energy accelerators in Dubna, Russia. Landauer Corp, Glenwood, Illinois, read the TLD detectors.

Results from these measurements are provided in ref. [8] and are summarized Table II below:

**Table II: Summary of Alpha/Proton and X-ray Measurements**

| Sample | Pd(2000A)/Glass | Pd(4000A)/Alumina | NiOx/Alumina |
|---|---|---|---|
| Exposure [hrs] | 48 | 118 | 118 |
| Sample history | Pd-coatingfractured. Eex ~ 0 | Survived, Excess heat, Eex ~ 20 % | No electro. at Ni-surface, Eex ~ 0 |
| $\langle N_\alpha \rangle \times 10^{-4}$, [$cm^{-2}$/s] | 4.8 ± 1.0 | 6.9 ± 1.2 | 0.0 |
| $\langle N_p \rangle \times 10^{-4}$, [$cm^{-2}$/s] | 28.6 ± 4.1 | 16.0 ± 0.2 | 0.0 |
| $\langle N_p \rangle / \langle N_\alpha \rangle$ | 6.0 | 2.3 | -- |
| X-ray, dose [mrad], TLD, $Al_2O_3$:C | 0.5± 0.5 | 0.2 ±1.0 | 0.1 ± 1.0 |

As seen, charged-particle emission was typically ~ $10^{-4}$ part./s-cm*$^2$, consisting of energetic protons clustered around ~ 1.7 MeV plus several bands of alpha particles grouped around 15 MeV. Emission was only observed from electrodes where significant electrolytic loading occurred. The ratio of alpha to proton rates increased from about 1:6 for partly cracked films with marginal excess heat (low loading) to 1:2.3 for stable films that produced significant excess heat. X-ray production was simultaneously observed, but with intensity only marginally above the initial TLD background.

This observation of MeV-particle emission data supplements the earlier SIMs/NAA measurement of multiple isotope production in the films [1-3], adding proof of nuclear reaction involvement in the excess heat production. The very low intensity of x-ray production is also consistent with the earlier measurements that required lengthy post-run exposures of the electrodes using high-speed photographic film [9].

## 6. SUPPORTING BASIC STUDIES

In addition to the electrode studies described above, various supporting experiments and theoretical studies are in progress. These include:

- New effects at ultra-high loadings (>0.9 proton/metal atom)
- Proton diffusion and dynamic loading in thin films (Devanathan cell method)
- Anomalous resistivity effects

This research has concentrated on extending the earlier deuterium-Pd wire loading experiment [10] to proton loading in thin films of the type described above. In addition, simulation studies have been undertaken to provide further insight into the phenomena involved at high proton loadings, especially as related to anomalous resistivity effects. The three characteristic loading regimes of interest are illustrated schematically in fig. 3.

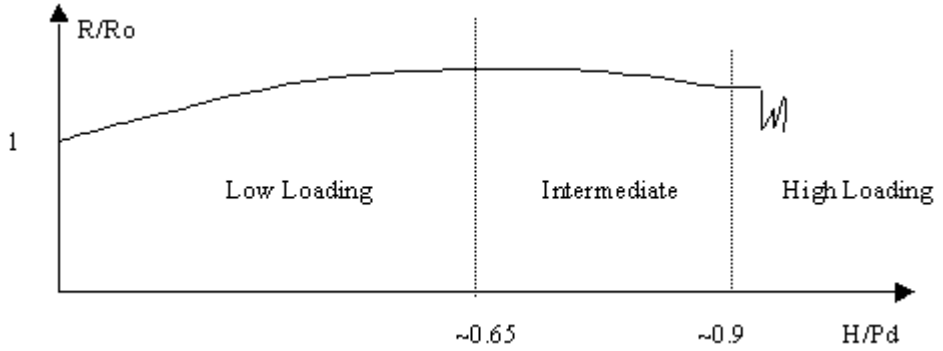


Fig. 3. Three Regimes in the $R/R_0$ vs. H/Pd Curve

This characteristic above has been well documented in the first two regimes for loading of deuterium or hydrogen in solid electrodes of Pd. In our earlier work the extension to the higher loading using Pd wires demonstrated a resistivity oscillation [10], in reasonable agreement with prior wire loading work at ENEA, Frascati. Our recent studies, described in [11,12], have extended this to proton loading of thin films. Concurrent lattice

structure simulation studies using the WIEN2k program indicate that, as proposed by others, the drop in resistivity at intermediate loadings is associated with the appearance of new band structures, giving in effect, parallel conduction paths. Based on these simulations, it is proposed [12] that the resistivity oscillation effect could be due to the formation of negative (time average) hydrogen sites associated with electron clustering in the vicinity of protons under ultra loading conditions. Additional theoretical studies are described in a companion paper [13].

## 7. POWER SCALE-UP POSSIBILITIES

While the absolute excess power in these studies is low (<0.5 W) due to the small amount of metal in the films, the specific power density, w/(g-pd), is 10 to 100 times that of the typical solid-electrode experiments. Thus a scale-up in power could be obtained using multiple electrodes as illustrated conceptually in fig. 4 to achieve a high packing fraction of Pd.

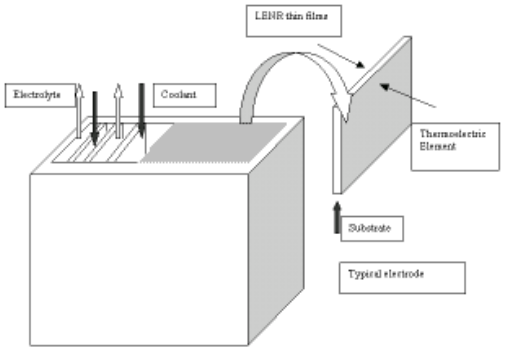


Fig. 4. Use of multiple electrodes to obtain a high packing fraction power cell.

While the present work has concentrated on wet electrolysis for loading of the films, gas pressure loading is also attractive for some applications. Thus a concept for achieving conditions with a hydrogen pressure method equivalent to the present electrolytic cells is shown schematically in ref. [14]. In this pressure loading design, hydride storage of the hydrogen is employed along with a Pd-black and semi-transparent layer technology to aid loading as $H^+$ and to minimize back diffusion, hence achieving high loadings and non-equilibrium proton conditions. Estimates of the performance of this cell [14] indicate a power-to-weight ratio that is very competitive with advanced fuel cell concepts.

## 8. CONCLUSIONS

In conclusion, the recent research described here has:

- Developed fabrication techniques for reproducible thin-film electrodes;
- Added support for earlier excess heat production measurements associated with ultra-high proton loadings in thin-film electrodes;
- Discovered MeV alpha-proton emission, supporting earlier reaction product evidence of nuclear origin of heat;
- Provided insight into highly loaded thin-film phenomena, including possible H- effect at ultra-high loading.

This research is continuing to study optimization of materials for maximum excess heat, lifetime, and minimum costs. Concepts have been present to scale the present single electrode experiments up to power cells with specific powers of order of 1 W/cc-cell-vol. using replication of electrodes in a closely packed multi-electrode cell. The ultimate objective is to achieve 100-W to 20-kW distributed power units. Still, the development of such units will require further improvements in electrode performance, combined with higher temperature operation and integration with a suitable energy converter.

**ACKNOWLEDGEMENTS**

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PowerPoint Slides From Lecture
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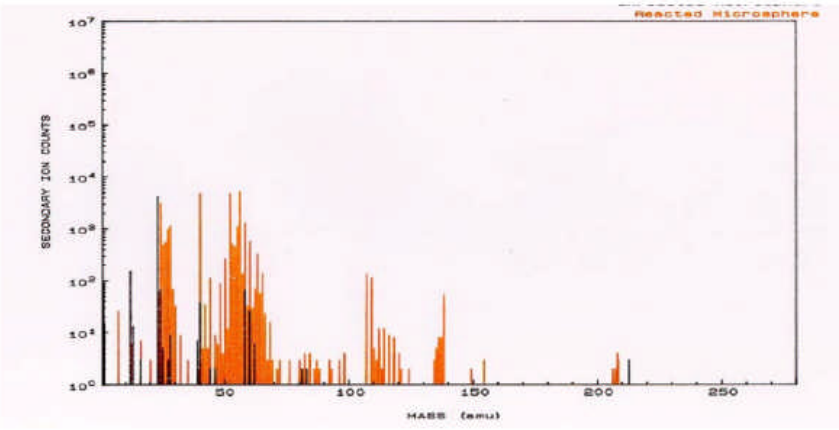
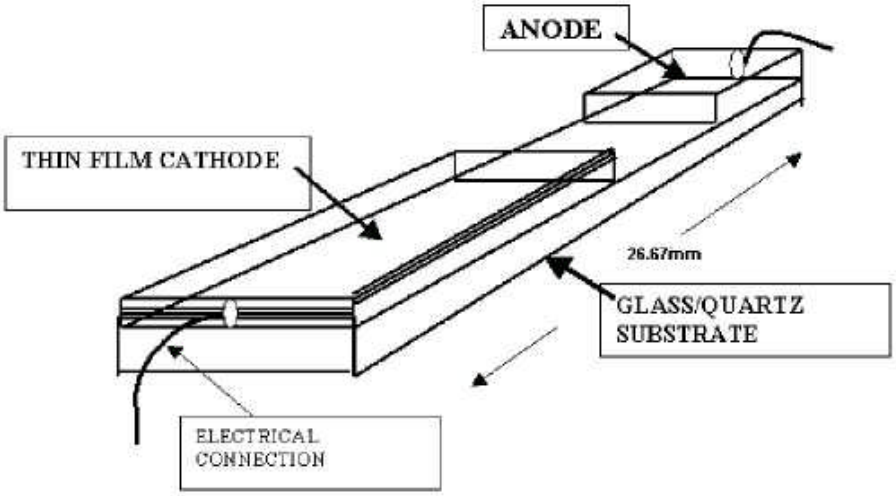
## LENR Reaction Product "SIGNATURE" from SIMS Measurement (ICCF-7). NOTE MASS "GROUPS"

INITIAL FILM HAS FEW IMPURITIES vs. RUN PRODUCTS.

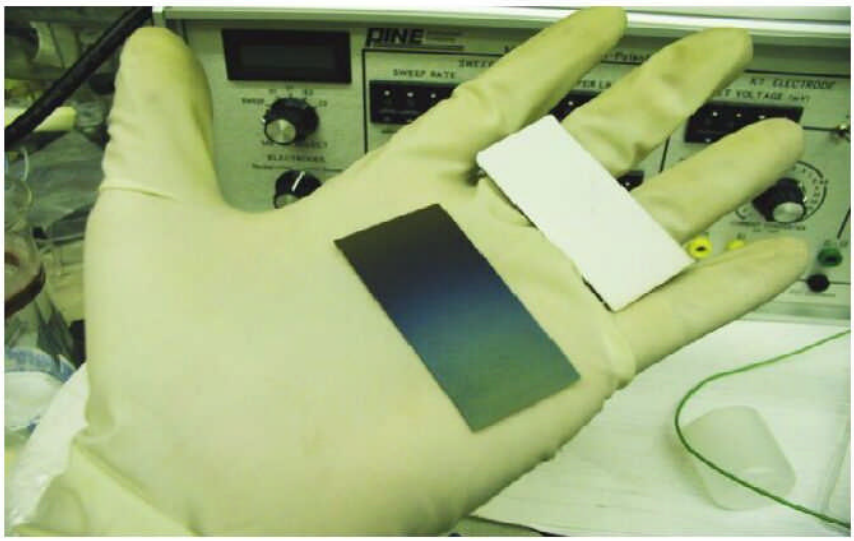
"PRODUCTION RATE" = (FINAL - INITIAL)



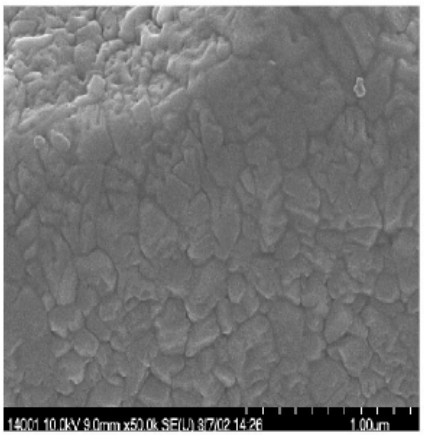
# "Building Block" Thin-Film Electrode

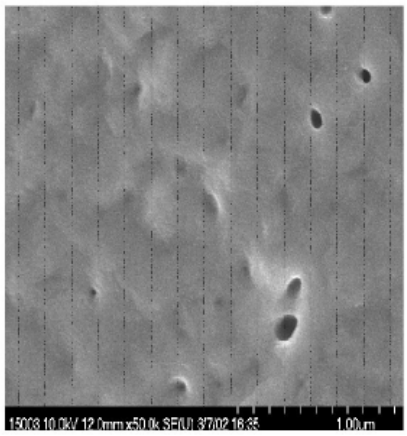



# Alumina Substrate Electrodes Serve as "Reference" Case



# Comparison of Film before (left) & after (right) Vacuum Annealing Pd Films





# X-ray pattern of $Al_2O_3$.

***Formation of Pd-Ni confirmed by Pd[1,1,1] ~ 40.178, Ni[1,1,1] ~ 44.582, Pd[2,0,0] ~ 46.733, Pd[2,2,0] ~ 68.241.***