

Bose-Einstein type D-cluster Electrode Development

G. H. Miley¹, X. Yang¹, H. Hora²

¹*Department of Nuclear, Plasma and Radiological Engineering, University of Illinois, Urbana, IL, 61801, USA*

²*Department of Theoretical Physics, Univ. of New South Wales, Sydney, Australia*

E-mail: ghmiley@illinois.edu

Abstract. Our recent research has developed a technique for imbedding ultra high density deuterium “clusters” (D cluster) in Palladium (Pd) thin film. Experiments have shown that in Pd these condensed matter state clusters approach metallic conditions, exhibiting super conducting properties. Using Temperature Programmed Desorption TPD system, the local concentration of hydrogen in the dislocation core is found to be $[H]/[Pd] \sim 1.8$. At near 70 °K Pd foil with abundant D clusters also show class II superconductivity, indicating the trapped hydrogen condensed into a metallic-like phase. Room temperature cluster formation is found to be adequate for the desired “nuclear reactive” sites. By careful calculation and experimentation, it is found the D cluster has a Bose Einstein Condensation state when a high deuteron diffusion flux is triggered. The resulting momentum transfer initiates reactions. The trigger can be electrochemical, giving LENR reactions. Thus this configuration can provide a small LENR power cell. Currently, we are searching for new methods to improve the volume density of dislocations. These methods mainly involve modifying Pd thin film with multi-layer structure.

1. Introduction

Recent SQUID measurement has shown ultra-dense states of deuterons with many more than 100 deuterons within a crystal defect in a palladium crystal are possible, and a superconductive state of these clusters was demonstrated in these experiments [1, 2]. Similar ultra-dense state of deuterons was seen at surface defects of iron oxide resulted in ion energies of 630 eV through Mass spectrometry measurements [3]. It may well be assumed that both cluster states are of the same nature though the states are concentrated at the surface in the iron oxide case due to the catalytic generation in contrast to the Pd samples with localization in the bulk volume [2]. In both cases their existence was confirmed by the LENR process [4] which likewise should be valid including when an inverted Rydberg state is present. Based on the excess heat measurements, local power densities exceeding a kW/cc are possible, promising very high energy density power units. Since the radiations emitted (protons, alphas, and x-rays) are not very penetrating (do not escape the cell structure) and no long lived radioactive reaction products are observed, LENR power units would be a remarkable “green” nuclear technology.

2. Earlier work on LENR at University of Illinois

There are a wide variety of fusion nuclear reactions. As illustrated in Figure 1, the original Pons-Fleishmann (P-F) reaction involved DD Fusion. But, instead of the normal reaction where the channel passes through deactivation of the He4 by transferring energy to the lattice which ultimately appears as heat, a number of researchers have reported transmutation reactions that involve interaction between deuterium/hydrogen and atoms in the host lattice, typically heavy metals. This branch is commonly termed Low Energy Nuclear Reactions (LENRs), although recently there has been a move to include DD reactions as LENRs as well. In this review we concentrate on host atom reactions.

		<u>D-D Reactions</u>	
			% branching
		hot fusion	"P-F" type
D-D	$T + p$	50	< 0.1
	$He-3 + n$	50	< 10^{-6}
	$He-4 + \gamma$	< 10^{-5}	99+
<u>LENRs</u>			
$p + metal \rightarrow products \text{ or } "fission" \text{ product array}$			

Fig. 1 - Comparison LENR reactions and the DD reactions.

Transmutation reactions can be broadly classified according to their products. Some experiments have observed a large array of reaction products with mass numbers ranging across the periodic table. These reactions are traced to multi-body events leading to a heavy compound nucleus, which can both decay, and fission into an array of elements [5]. The other set of experiments lead to one or few distinct isolated products [6]. These reactions may or may not involve multi-bodies but the net result is direct formation of the reaction products as opposed to the disintegration of a compound nucleus. Earlier works on transmutation reactions involved thin films on microspheres, while our recent studies at the U of I have converted it to thin films coated on a flat alumina substrate as illustrated conceptually in Figure 2. In this design, a double-layer Pd/Ni thin film, at 8000 Å and 1000 Å respectively, sputtered on ceramic Al₂O₃ substrate at rate 0.41 Å, was used as a cathode. The electrical current flow is parallel to the thin film surface so that a high current density and high proton flow rate are obtained along with a high deuterium density [7].

The possible heat evolution during electrolysis runs was detected by a high sensitivity open-type calorimeter in a fixed temperature of 28.0 ± 0.05 °C. [8] To check actual performance during Foreground electrolysis runs (with Alumina/Pd-Ni samples), including heat convection, bubbling, electrode's geometry, and positioning, as well as H₂+O₂ recombination, smooth Pt sheets were used as cathodes in Reference runs, since Pt does not produce excess heat in the light water electrolysis [9]. The performance of this runs at various current (50 ~ 600 mA) is defined by the calorimeter "heat recovery" value $R = P_{th}/I(U-U_0)$, where, P_{th} is the thermal power measured in calorimeter by thermistors; $I(U-U_0)$ is Joule heating P^* , in which I and U are the electrolysis current and voltage, respectively; U_0 is an effective water dissociation potential (WDP) (2.01 V for Pt/Pt pair and 2.06 V for Pd-Ni/Pt pair) [10]. A constant electrolysis current ranging from $I = 100 \sim 600$ mA ($j = 8.5 \sim 50.0$ mA/cm²) corresponding to a cell voltage U 3.5 ~ 5.5 V were applied to Alumina/Pd-Ni cathodes Typical kinetics of the heat measurement (P_{th} vs. elapsed time t) are presented in Fig. 3.

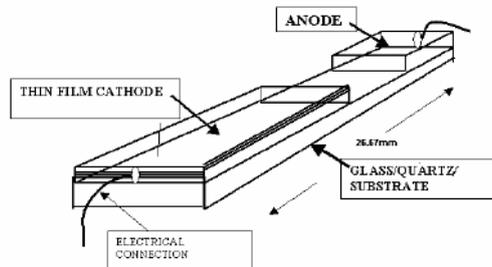


Fig. 2 - Recent Work Uses a Unique Integrated Thin film Plate-Type Electrode.

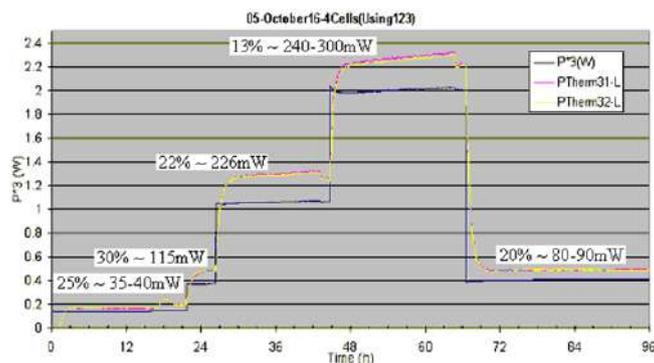


Fig. 3 - Heat measurement for two layers electrode: 8000Å Pd and 1000Å Ni on Alumina. Correlation between Transmutation Products and Excess Heat

As seen, shortly after starting the run, the thermal yield P_{th} exceeds Joule heating P^* . The current was run in “jumps” purposely to examine changes as a function of input power. The maximal heat recovery R of 135 % occurs at electrolysis current $I=100$ mA ($j = 8.0$ mA/cm²), while the corresponding absolute value of excess heat production is $W_{ex} = 60 \pm 28$ mW. Increasing the current density to 48 mA/cm² leads to decrease in heat recovery R from 135 to 115 %. In typical cases the excesses heat evolution from the cathode at $I = 200$ mA continues for ~ 15 days. The decrease in R during a long Foreground run is accompanied by an increase in cell voltage. The last process indicates an increase in the cell inner resistance due to defects and micro-cracks generation in Pd/Ni cathode. The cathode samples survive, i.e. they do not detach the Alumina substrate during these long runs. Thermistor calibration runs repeated immediately after this Foreground run show that calorimeter parameters, (including temperature change vs. input power) did not change during the time of operation with Alumina/Pd-Ni sample (15 days). This gives a proof of stability of the calorimetry system and indicates that our measurements of excess heat production were actually correct.

Although good progress has been made on direct correlating He-4 production and excess heat, supporting the D-D reaction hypothesis, the correlation between the transmutation and excess heat production, as well as the product measurements, still leave open a fairly large error band. Earlier, we have observed a variety of reaction products (isotopes) with masses both higher and lower than that of the host electrode material in thin film electrodes, [11][12][13], suggesting the existence of proton-metal initiated reactions in such LENR cells. Later, we discussed evidence that the production of these reaction products is correlated with the excess heat described above [14]. Such a correlation for LENR reactions would be equivalent, in principle, to the correlation of He-4 with excess heat that is reported for heavy water-Pd experiments where a D-D reaction is postulated.

A characteristic result, shown in Fig. 4, indicates that a variety of reaction products occur with masses lying well below and above that of the base electrode metal (Ni and Pd in Fig. 4, of cf. numbers defined in the references). A striking pattern consistently observed in these measurements is that the high-yield reaction products occur in four mass ranges, roughly $A = 20-30$, $50-80$, $110-130$, and $190-210$ [12] [13]. The high-yield products ($> 10^{13}$ atoms/cc-sec in Fig. 4) are well above background impurity limits, but some uncertainties due to possible impurities, especially in the electrolyte, still plague measurements of the lower yield products ($< 10^{13}$ atoms/cc-sec in Fig. 4). Interestingly, a recent study by Widom and Larsen claimed that the theoretical transmutation calculations reasonably well match the Miley & Patterson result (see fig. 3 in [15]).

Two other points should be stressed. First, no neutrons have been observed in these experiments despite repeated attempts with sensitive detectors Likewise, no tritium has been observed in electrolyte analysis after runs. [Note: some LENR experiments run other quite different conditions have reported neutrons and tritium production. Thus, depending on the experimental design, there appear to be different branches to the LENR reactions]. Second, as noted in Miley and Patterson [11], long exposure of photographic films to electrodes removed after extensive runs indicate low levels of soft x-ray and/or beta particle emission.

This is consistent with the recent theoretical models where the fission-like process occurring in the transmutations has little excess energy. Thus, unlike uranium fission where the intermediate compound nucleus is highly excited, this energetically “soft” type fission leads to many products that are stable and others only slightly unstable [15] [16]. This process then avoids energetic gamma emission and the production of highly radioactive nuclear waste encountered in uranium fission systems. These remarkable results are basic to this new field of LENR nuclear physics and the unique processes that can be created by causing nuclear reactions in condensed matter. Please note that the “production rates” shown represent a time average over the experiment run-time since it was only possible to take samples at the beginning and end of a run. Likewise, the excess heat reported represents an average over the run, so that a comparison of these two results is consistent. Thus, to test a possible correlation between transmutation products and excess heat, the measured product yields are used along with their respective binding energies to compute a theoretical “excess power”, W_{ex} , as shown in Fig. 5a. The computation is straightforward but tedious due to the large number of reaction products produced. Basically, W_{ex} is computed by taking the product of all the isotope yield rates times their binding energies and subtracting the corresponding product for the “fuel”. A key point is the determination of the amount of original material that is consumed or “burned up.” This calculation is done by first allotting the maximum number of reaction product nucleons to the metal nucleons. Then, any remaining nucleons are attributed to the protons, allowing for a variable proton/metal atom ratio to retain generality. This balance rests on the assumption that the protons plus the electrode metal (e.g. Ni in this case) are the reactants in LENR cells. This follows because protons in the light water electrolyte cannot react with themselves. The ‘salt’ employed in the electrolyte, e.g. Li_2SO_4 , should not be involved in the reaction either, because various workers have used different salts while still obtaining similar reaction products.

Product yield results from three runs (run numbers refer to experiments described in Miley and Patterson (1996) where adequate information was available for this type of evaluation are summarized in Fig. 5b. Although one has a mean measured value that is a factor of two larger than the calculated value, two of the results show quite close agreement. While these results are not definitive, in view of the many uncertainties in both of the calculated values (due to uncertainties in the yield measurements) and in the calorimetry, the agreement obtained strongly suggests a relation between products and excess heat. The situation where heavy water is used instead of light water, as reported in some other LENR studies (see [17]), is less clear but again appears to involve proton-metal reactions. In that case, p-metal reactions could occur simultaneously with D-D reactions. More study is needed to resolve possible reactions involved this important regime.

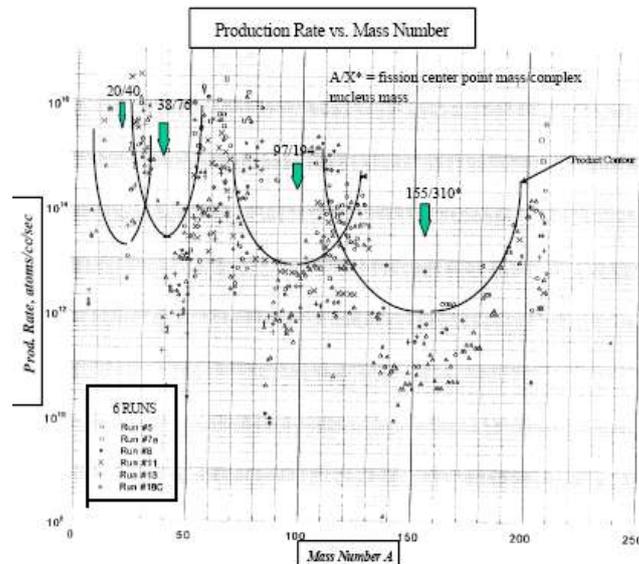


Fig. 4 - Reaction Product Yield vs. Mass Curve.

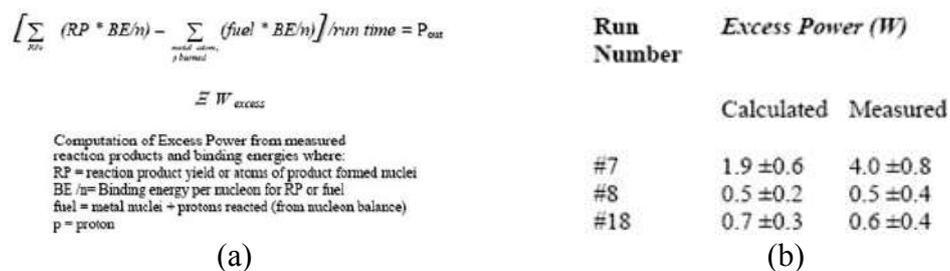


Fig. 5 - (a) Computation of excess power from reaction n product measurements. (b) Results from Energy Balance Calculations for Three earlier Thin-Film experiments. All experiments used Li₂SO₄ in H₂O for the electrolyte and thin-film Ni coated cathodes.

Another way of viewing these data is to calculate the energy released (the observed excess heat times the run time) and divide by the number of Ni atoms reacted (based again on the number of nucleons associated with the measured quantities of reaction products observed). Then, for the runs referred in Fig. 5b, an energy release of order of 150 keV/Ni atom reacted is obtained. This value is consistent with nuclear as opposed to chemical processes. It is several orders of magnitude less than the energy released in neutron-induced fission, but is roughly in the range of “soft” fission releases predicted for LENR conditions [16].

These results also bear on an issue that is often raised about the LENR experiments: how can a positive excess power occur since the base metal involved, such as Ni, has a binding energy per nucleon near the peak of the binding energy-mass curve? In the present analysis this can be explained by noting that the “fuel” i.e. the reactants, are a mixture of protons and metal. Then the average binding energy of the reactants (p + metal) is reduced below that of the metal alone. As a result, there is an expanded range of reaction product masses laying around the mass of the base metal that offer a positive energy release, i.e. a positive Q-value for the reaction. Still, the fact that some reaction products lie outside of this range might seem to infer that reactions occur despite a negative Q-value, but then a very large input energy would be needed to drive the reactions. This dilemma is overcome, however, if the reaction occurs through multi-step excitation and/or formation of a compound nucleus which can split up or fission into a variety of reaction products of different masses [11] [16]. The energy balance requirement is that the formation energy of the compound nucleus must be supplied. Subsequently, the break-up energy is, in effect, shared among products.

In order to obtain further proof for nuclear reactions in the thin-film cathodes [18], the detection of nuclear radiation accompanying the electrochemical loading of those cathodes is strongly desirable. Although the studies of long-range alpha emission have been done after the electrochemical loading of Pd with deuterium/hydrogen [19], it was a great interest to expand the experiments on charged particle detection to in-situ measurements during the electrolysis process. Unfortunately, it is hard to apply electronic SSB and X-ray detectors directly to the cathode during electrolysis experiment. Thus, a technique using non-electronic detectors (CR-39 and thermal luminescence detector (TLD)) was employed to allow in-situ measurement of energetic charged particles and X-rays during the electrochemical loading of the flat-plate Pd-thin film cathodes.

In the electrolysis experiments the freshly opened CR-39 detector chips or TLD crystal were attached either to the Pd thin-film cathode (Foreground) or to the substrate side or/and immersed in electrolyte in the cell (Background). The charged particle and x-ray detection were carried out simultaneously with excess heat measurements in open type calorimeter. The electrolysis current and duration during one Foreground run were normally varied in the range of 50-400 mA and 2-30 days, respectively.

Fig. 6 and insets show the Foreground and Background CR-39 reading for both alpha particles and protons. The count rates of protons and alphas after Background subtracting are statistically significant with Background level being close to zero. In the control experiment with CR-39 detector attached to the thin film NiO_x (obtained by annealing of Alumina/Ni(4000A) sample in air atmosphere), where despite of the high voltage applied (U~10.0 V), the electrolysis current in the cell was very low (I~1.0 mA), and no tracks with d < 7.8 μm were detected.

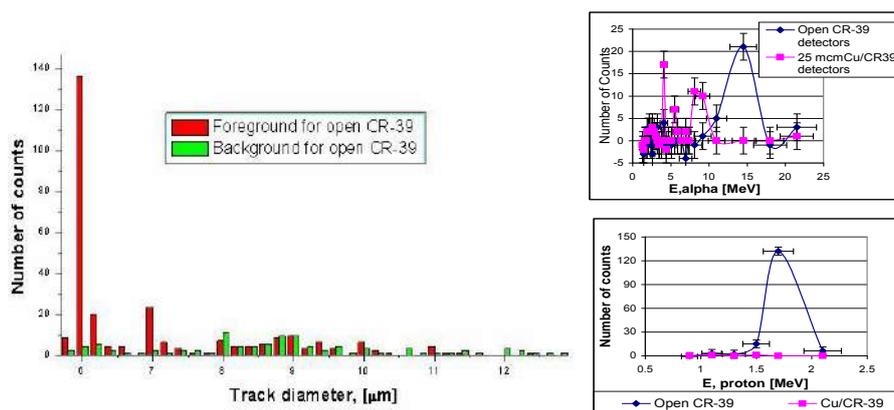


Fig. 6 - Electrochemical loading of Pd thin film cathodes on dielectric substrates unambiguously produced high-energy charged particles: 1.5-1.7 MeV protons and 11-16 MeV alphas. Insets: Alpha-tracks energy distribution and Proton energy distribution after background subtracting.

In contrast to charged particle detection results, the statistical significant level and reproducibility of X-ray emission was not satisfied. Some result (e.g. Pd/Glass sample with LIF TLD) showed consistency between the emitted X-ray fluency and absorbed dose but others (e.g. other cathode with $\text{Al}_2\text{O}_3:\text{C}$ TLD) did not show statistically significant results due to higher initial background level (~ 10 mrad) of these TLD compared to that for LiF. The X-ray measurement results indicate the absence of massive nuclear reactions caused by nuclear transmutation in Pd [18]. However, these results could not rule out the possibility of a weak X-ray emission as a result of the charged particle generation or Pd thin film fracture or detachment from the dielectric substrate.

In summary, the observation of MeV particle emission, combined with the transmutations and excess heat measurements, provide extremely strong evidence for nuclear reactions occurring in the thin films during electrolysis. These results, combined with recent observations of localized sites (“clusters”, discussed next) have lead us to consider the new approach to power producing LENR cells described here.

3. Ultra High Density Deuterium Cluster Electrode Fabrication

The SQUID magnetic measurements described in Lipson *et al.* [1] show “clusters” have characteristics of a type- II superconductor. Cluster regions can have hydrogen densities approaching $10^{24}/\text{cc}$ (See Fig. 7). Dislocation loop cluster type electrodes are fabricated by cyclic loading-unloading, hence being named “Dislocation Loops by Repetitive Loading-DeLoading (DLRLD)” electrodes. These DLRLD electrodes are based on studies where high loadings in dislocation loops in treated Pd have exhibited properties associated with a superconducting phase termed a “cluster”.

After several loading-unloading cyclings, the Pd/PdO: H_x was annealed at 300 °C for 2 hr to remove all the weakly bond hydrogen or deuterium. Then, a high-vacuum thermal desorption technique is used to estimate residual hydrogen concentration in the Pd/PdO: H_x samples (Fig. 7C). The samples were heated in a high vacuum (10^{-8} Torr) chamber with a quadruple mass-spectrometer. The hydrogen desorption peak area and the temperature of its maximum were found by analyzing the desorption species and comparing their yields to background data from the Pd/PdO : H_x or Pd/PdO).

By using Garlic-Gibson kinetics model, the activation energy of desorption is found to be approximately 0.65 ± 0.10 eV, which is consistent with the result of Kirchheim for hydrogen trapping at dislocation core sites in cycle Pd. This indicates that the hydrogen is solely bound inside the deepest core sites [approximately one Burgers vector (2.75 \AA)], meaning that all residual hydrogen is localized inside the dislocation loops (in the direction of Pd) determined by Burgers vector $b [101] = 2.75 \text{ \AA}$. Then, the dislocation density calculation give x_{eff} is ~ 1.8 , suggesting superstoichiometric hydride formation in the deep dislocation cores.

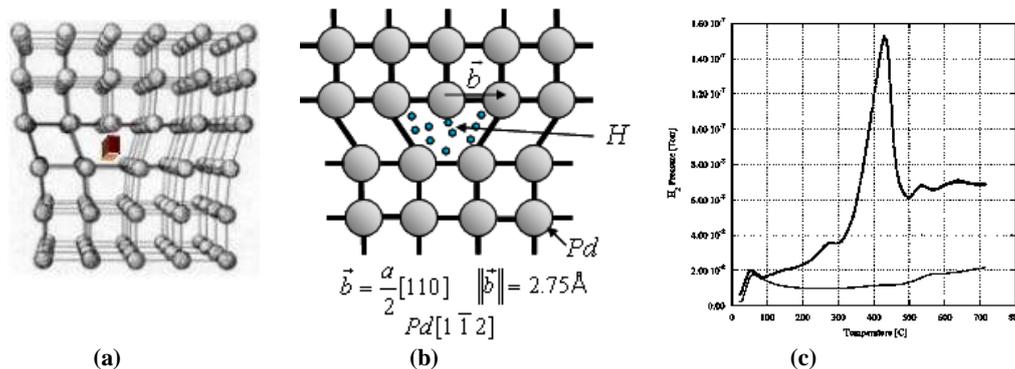


Fig. 7 - SQUID magnetic measurements show clusters have characteristics of a type-II superconductor. Cluster regions can have hydrogen densities approaching $10^{24}/\text{cc}$. (a) 3D scheme of typical edge dislocation core (see orange bar) in the Pd crystal. (b) Cross-section of A. (c) Thermal desorption measurements of Pd/PdO : H_x (thick solid line) and Pd/PdO (thin solid line).

4. Other Ultra-High-Density Deuterium Cluster – Rydberg Matter

Encouraged about the result [1, 2] that SQUID measurements confirmed the localization of more than 100 nuclei of hydrogen in a crystal defect within a palladium crystal, an estimation was given [20] how many such defects may be generated in a crystal. A density of such defects in crystals may well be more than 10^{19} cm^{-3} before the crystal is breaking. This is the experience from the property in silicon crystals where such densities of doped atoms are well known. Similar density of defects with voids has been produced in silicon too by very intense electron bombardment. [21, 22, 23, 24] If these voids are then in an average distance of 10 atoms in the crystals for laser fusion targets based on LiH or similar crystals at room temperature, the targets with a density of fusion fuel of 1000 times solid state density may be possible [b].

A necessary condition, however, is that the clusters in the crystal volume should have the density of 10^{24} cm^{-3} . These densities have been confirmed [3] from clusters which were produced in crystal defects at the surface of iron oxide by catalytic processes of inverted Rydberg states. These densities were proved from the Coulomb repulsion energy of deuterons in such states as time-of-flight measurements showed 630 eV w. This corresponds to a distance of the deuterons of 2.3 pm within the inverted Rydberg clusters. The repulsion process is given, when laser irradiation of 546 nm wave length removes the neutralizing electrons between the deuterons [3].

The mechanism of the electron emission by the laser radiation has been analyzed by inclusion of the correspondence principle of electromagnetic interaction [25]. A virtual quiver motion of the electrons in the quantum state in the inverted Rydberg cluster arrives at a quiver elongation of 2.3 pm at a laser intensity of $1.01 \times 10^{10} \text{ W/cm}^2$. This intensity was just measured as the lowest threshold [3] for the measurement of the 630 eV deuterons.

A further conclusion of this analysis is that the state of the deuterons in these clusters represents an effective Bose-Einstein condensation at room temperature. [26] A further result is for a hydrogen atom, the virtual quiver motion of the electron within the laser field with an elongation of the Bohr radius corresponds to the well known ionization energy of 13.6 eV. This energy has the unique value of $\alpha^2 mc^2/2$ expressed by the fine structure constant α and the rest mass energy of the electrons mc^2 with the electron mass m and the vacuum speed of light c .

5. Conclusion

This paper makes a strong case that condensed matter deuterium cluster formed in dislocation loops are one way to achieve nuclear reactive sites in LENR electrodes. The problem to date has been that the volumetric density of such sites, which is highly reactive, have a low density of sites per unit volume.

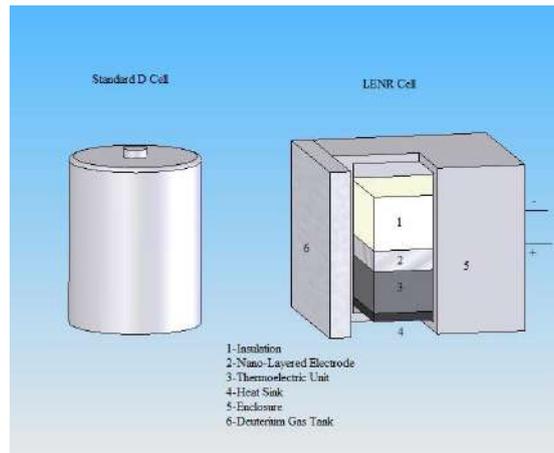


Fig. 8 - Small LENR Battery -- design based on present experimental data base

Possible methods to achieve a high volumetric density of sites (i.e. achieve a massive cluster electrode (MCE) that are under study are briefly outlined. This is thought to offer an orderly “roadmap” for moving on to very unique future power cells for both space and commercial use. Such cells would employ nuclear reactions for energy but enjoy no neutron release and a minimal radioactive product buildup. These characteristics are based on the experimental observations cited here where any neutron levels were below detection limits; MeV particles, while observed, had very low emission rates; and the reaction (transmutation) products created are generally close to stable, with only soft x-ray or beta emission being observed.

Based on the excess heat measurements, local power densities exceeding a kW/cc are possible, promising very high energy density power units. Since the radiations emitted (protons, alphas, and x-rays) are not very penetrating (do not escape the cell structure) and no long lived radioactive reaction products are observed, LENR power units would be a remarkable “green” nuclear technology. A sketch of a small D-cell equivalent LENR battery is shown in Fig. 8. The cell shown has a deuterium gas “fuel” tank attached.

Gas loading is used rather than electrolysis for compactness. Heat flow is directed to the outer casing through a thermoelectric element using an insulation and heat sink design. Modular sections connected in series allow a 1.5 V output at 0.1 A. This type of battery must be used in devices where natural convection air cooling or other heat flow dissipates heat from the battery casing. The battery run time is determined by the amount of deuterium stored refilling the depleted tank is provided by pump down and gas injection through a filtered line connected to a “filling station”. The unit shown is designed for 1000 Ahr per gas fill. The main technological step needed before construction of this battery is to finish development of the nano-layered electrode structure described earlier.

6. References

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