Cold Fusion by Gas Loading: A Review

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Abstract
Since the announcement of the discovery of Cold Fusion by Fleischmann and Pons in 1989, scientists developed techniques other than electrolysis in order to achieve the Cold Fusion effect. A lot of effort has been made to develop gas loading. This approach has been followed by a large number of scientists all over the world and has proven to be productive. Excess heat has been demonstrated, as well as production of helium, tritium, neutrons, gamma rays and transmutation. Not only the palladium-deuterium system has been explored, but also titanium-deuterium and nickel-hydrogen, as well as several other systems. In this paper we review the large body of work that has been accomplished so far.

1 - Introduction
At the public announcement of Cold Fusion on March 23, 1989 by Pons and Fleischmann (1), the world discovered that there was a possibility of an endless energy source. Using an electrochemical method, they showed that it is very likely that nuclear reactions can occur at low energy. Interestingly, as early as 1986, Kluev et al. (2) had shown the production of neutrons when perovskyte type compounds loaded with deuterium passed through the ferroelectric transition temperature. They also showed neutron production when heavy ice was crushed (3). These early experiments indicated that nuclear reactions could occur at low energy. Following the press conference most of the experimental work involved electrolysis. However, a large number of scientists started experimenting with gas phase materials. It is the purpose of this paper to review the large quantity of data available in this field.

In this paper we do not cover interesting works done with solid-state electrolytes and plasma discharges. Even though these experiments are performed in gas phase and not in liquid phase, there are many papers about it, which deserve review papers of their own.

2 – Why electrochemistry?
Fleischman and Pons (1) decided to perform their experiments in an electrolytic cell. The main reason being that according to thermodynamics, using Nernst law it is possible to achieve very high-pressures in the cathode of an electrochemical cell. At low pressure, in the case of hydrogen loading, the relation between the over-voltage and the pressure or fugacity inside the cathode is given in equation (1).

\[ V = \frac{RT}{nF} \log \frac{P}{P_0} \] (1)
Where $V$, is the over-voltage, $R$, the perfect gas constant, $T$, the absolute temperature, $n$, the number of electrons involved in the water dissociation, $F$, the Faraday constant, $P$, the pressure or fugacity inside the palladium cathode and $P_0$, the atmospheric pressure.

This formula indicates that if one can generate large over-voltages on the cathode, this will correspond to loading of the cathode with a very high pressure of deuterium gas. However, the challenge is to generate these over-voltages. There are several ways of doing so, one of them being an increase of the electrical current density. It is clear that achieving such high pressures in gas phase is impossible with our current technologies. That is one of the reasons the electrochemical method was selected. Another one is the simplicity of it, and the relative ease of doing calorimetry with high precision. It is also easy to change the current and therefore the conditions of the electrolysis.

3 – Why gas loading?

Even though electrochemistry has many advantages as explained above, it also has several limitations:

- It is difficult to achieve large over-voltages, because either it is necessary to have high current densities or to deposit a specific material at the surface of the cathode that helps produce them.
- It is difficult to maintain very clean experiments because many impurities in the electrolyte will end up on the cathode surface by cathodic deposition. Even if the concentration of impurities is low, very small amounts will produce atomic level layers of deposits at the cathode surface capable of greatly modifying the surface reactivity.
- Analysis of by-products of the reaction such as helium, alpha particles, transmutation is easier in gas phase.
- As pointed out by Fleischmann and Pons (4), there is a positive feedback, and excess heat increases with temperature. With water electrolysis, we are limited to 100°C at atmospheric pressure. It is possible to work at higher temperatures, but this calls for the use of pressurized cells or molten salts, which are much more difficult to operate.
- Finally it seems that future technological applications will be difficult to develop with electrochemical cells.

There are definite advantages to gas loading:

- Pressures are easily controlled.
- It is possible to maintain clean environments.
- Operational temperatures can be high.
- It is easier to measure helium and charged particles.
- There is more potential for practical applications.

There are also some disadvantages:
It is difficult to achieve very high pressures.

To do good calorimetry requires more expensive equipment with this technique than with electrolysis.

4- Excess heat production

Excess heat is certainly the goal of many scientists in the field of Condensed Matter Nuclear Science, because it would result in a practically infinite source of energy. (At present energy consumption rates, deuterium fuel will last for billions of years.) Therefore many different ways have been developed to reach that goal. In the following sections it is shown a number of different ways to succeed.

4.1 Palladium tubes and membranes

As early as 1989, Fralick et al. (5) at NASA loaded deuterium in a hydrogen purifier at 370°C and a pressure of deuterium gas of 1.4 MPa on both sides of the purifier palladium tube. Their primary goal was to detect neutrons. When the deuterium was pumped out, a temperature rise was observed. When filled with hydrogen no temperature variation was detected. However no neutrons were observed above background. This was a very simple and convincing experiment showing anomalous excess heat in the palladium deuterium gas system.

Many other experiments have been performed since then with many variations: solid metals, thin wires, and powders. Romodanov et al. (6) have shown the production of excess heat when deuterium flows through palladium foils. Similar results have been observed by Li et al. (7-14). In a similar type of experiments, Tian et al. (15) detected heat after death. Manduchi et al. (16) observed anomalous effects when palladium is cooled from 900°C to room temperature.

4.2 Palladium powders

Palladium powder loads deuterium better than solid palladium. This is why several groups have used such substrates to detect excess heat. Kirkinskii et al. (17,18) have measured one Watt of excess heat per gram of palladium in gas-loaded experiments. However, when palladium powder is heated, sintering occurs, and the original particle size is destroyed. Therefore it is better to use dispersed palladium in a medium. Case (19,20) tried various palladium catalysts and finally found one that was effective in producing excess heat. McKubre et al. (21) successfully replicated this experiment. The work by Arata and Zhang (22-38) is of particular interest. They started with a unique design using a Double Structure cathode consisting of a hollow palladium tube filled with palladium black. Even though their first set of experiments used electrochemistry, this was only used to dissociate heavy water and purify the deuterium gas at the outside wall of a solid palladium tube. Only deuterium reached the nanoparticles in active portion of the experiment, on the inside of the cell. The deuterium diffuses through the walls of the tube and finally enters the center of the tube as atomic/molecular. Therefore this can be considered a method of performing gas loading of palladium black. Later, Arata and Zhang (39-42) used another approach. They loaded palladium nano-powder embedded in a ZrO₂ matrix by high deuterium pressure. They obtained very high loadings, and excess heat even at room temperature (40). Marmigi et al. (43) showed anomalous excess heat
using an oxidized palladium wire. Celani et al. (44) showed high loading and large excess heat when thin palladium wires are coated with nano-particles.

4.3 Excitation
The reaction has been triggered by various means of excitation. Li et al. (45) have measured excess heat when RF heating was used. Nassisi et al. (46-48) observed transmutations when irradiating etched palladium samples with an excimer laser.

4.4 Out-diffusion
One of the major goals of research in Condensed Matter Nuclear Science is to determine the mechanism responsible for the anomalous observations. It has been assumed that the reactions are not located in the bulk of the material, but at the surface of the metal. Early on Yamaguchi et al (49,50) have observed excess heat when deuterium flows out of a palladium foil covered on one side with gold. Similarly Liu et al. (51) show that excess heat occurs when deuterium flows out of the palladium. The amount of excess heat increases with the deuterium flow. Narita et al. (52) have also shown heat production when hydrogen is pumped out of a palladium foil covered on one side with MnOx or gold. Lipson et al. (53) showed excess heat while de-loading a palladium foil oxidized on one side and covered with gold on the other. Biberian and Armanet (101,102) showed excess heat production when deuterium flows out of a palladium tube.

4.4 Palladium wires
Li et al. (54) observed excess heat from palladium wires in deuterium gas. Marmigi et al. (43) have shown anomalous excess heat production when a previously oxidized palladium wire is heated in a hydrogen atmosphere. Celani et al. (44) showed that extremely large excess heat were detected when long (60 cm in the experiment shown) and thin (50 μm) Pd wires, coated with nano-materials, underwent the phase transition from beta to alpha phases and large current densities (10-50 kA/cm²) were applied along the length of the wire. This effect was stable over time. The power density was up to 400 W/g of palladium. The operating temperatures where as large as 300-500°C. The effect happened mainly in pressurized deuterium atmosphere, however an effect was also observed at much lower levels using hydrogen.

5 – Neutron production
As early as 1986, Kluev et al. (2,3) discovered the production of neutrons when a strong mechanical action is exerted against heavy ice or LiD. Later, Derjaguin et al. (55) showed that neutrons were produced when titanium deuteride chips were subject to mechanical vibrations. Jin et al. (56) observed neutrons in a YBa₂Cu₃O₇ sample loaded with deuterium. Shioe et al. (57) and Shirakawa et al. (58) similarly observed neutrons in LiNbO₃ fractured in a deuterium atmosphere. Aiello et al. (59) have also observed neutrons in palladium loaded with deuterium. De Nino et al. (60-62) detected neutrons while varying the temperature of titanium foils loaded with deuterium. Menlove et al. (63-66) showed similar results. Fabrizio et al. (67) also found neutrons the same way. At the same time, Claytor et al. (68-70) and Tuggle et al. (71) at the Los Alamos National Laboratory detected neutrons by pulsing currents with palladium electrodes in deuterium gas. Since then many other scientists have tried to detect neutrons in gas loading experiments. Bressani et al. (72-74) have measured neutrons emission when
titanium foils are loaded with deuterium. Yamaguchi et al. (49,75) have demonstrated production of neutrons when deuterium de-loads from a palladium foil loaded with deuterium with one face of the palladium covered with gold. Shirakawa et al. (76) showed production of neutrons when LiNbO$_3$ is fractured in a deuterium atmosphere. Iwamura et al. (77) showed production of neutrons when a palladium foil loaded with deuterium with one face of the palladium covered with gold. Shirakawa et al. (76) showed production of neutrons when LiNbO$_3$ is fractured in a deuterium atmosphere. Iwamura et al. (77) showed production of neutrons when a palladium foil loaded with deuterium is heated up. However Garg et al. (78) in a similar experiment with a palladium wire loaded with deuterium or hydrogen did not detect any neutrons. Aoki et al. (79) could not detect any neutrons above background in a tungsten sodium bronze loaded with deuterium when loading or de-loading the material. Shinojima et al. (80) did not detect neutrons when flowing deuterium through a palladium foil. However Lipson et al. (53,81) and Roussetski (82) did detect neutrons during exothermic desorption using a Au/Pd/PdO:D sample. Also, Dougar-Jabon et al. (83) detected neutrons during alpha/beta phase transition in palladium deuteride. Chicea et al. (84) detected neutrons in titanium deuteride when the metal was loaded at high temperature then the temperature was lowered. Itoh et al. (85) observed neutron emission when out-gazing a highly loaded palladium foil.

6 – Tritium detection

As early as 1990, Iyengar et al. (86), Srinavasan et al. (87) Iyengar et al. (88), Kauskik et al. (89) and Rout et al. (90,91) have shown production of tritium in palladium and titanium samples loaded with deuterium. Lamza et al. (93) measured tritium in various metals loaded with deuterium. De Nino et al. (62) detected tritium in titanium loaded with deuterium. Yamada et al. (94) have shown the production of tritium when deuterium is pumped out of a palladium foil covered on one side with MnO$_x$. Narita et al. (52) have observed mass three corresponding either to tritium or helium-3 when hydrogen is pumped out of a palladium foil covered on one side with MnO$_x$ or gold. Similarly Wei et al. (95) have also observed mass three when deuterium flows through a palladium foil. Claytor et al. (70,97) measured tritium with Pd-Si electrodes in deuterium gas. Clarke et al. (96) detected tritium in titanium loaded with deuterium and later (98) observed production of tritium in a cell similar to the one of Arata. Romodanov et al. (6,99) have also detected tritium. Lipson et al. (100) detected significant amounts of tritium when cooling YBa$_2$Cu$_3$O$_{7-D_x}$, to its Curie temperature (88-93K) in deuterium gas. Heating a palladium foil loaded with deuterium by Iwamura et al. (77) showed production of neutrons.

7 – Helium measurements

Detection of helium-4 is important in Condensed Matter Nuclear Science. Its measurement is another proof of the nuclear origin of the phenomenon. However few experiments have been performed so far. Botta et al. (103,104) have detected helium-4 using palladium foils covered on one side with gold and loaded with deuterium. Clarke et al. (96) detected tritium in titanium loaded with deuterium. Qiao et al. (105,106) showed production of helium from palladium in deuterium gas. McKubre et al. (21) have detected helium-4 in a Case type experiment, and showed a correlation between helium-4 production and excess heat. Arata and Zhang (32,107-109) showed, production of helium-3 and helium-4. Clarke et al. (98) showed
the presence of helium-4 and very large quantities of helium-3 in a cell similar to the one
developed by Arata.

8 – Charged particles
Mo et al. (110,111), Wang et al. (112), Jin et al. (113), Li et al. (114) and Dong et al. (115)
showed evidence of charged particles emission from deuterated palladium. Cecil et al. (116)
have observed charged particles in deuterated titanium. By a coincidence method Jones et al.
(117,118) showed that energetic protons and tritons are produced in non-equilibrium conditions
in titanium deuteride samples. Roussetski et al. (119) have also detected charged particles from
titanium hydrides and deuterides triggered by a pico-second powerful laser beam. Cecil et al.
(120) have also measured charged particles from palladium deuterides. Lipson et al. (121)
oberved beta emission from deuterated titanium.

9 – Loading experiments
We have shown above that Nernst law explains why it is possible to load hydrogen or
deuterium at high levels by electrolysis in palladium or other metals. In gas phase experiments,
it seems that the limitation comes from the difficulty in reaching high pressures of gas.
However, several researchers have tried to load palladium and other metals by various ways.
Huang et al. (122) have shown that it is easier to load hydrogen than deuterium in palladium.
They also showed that pulse heating a palladium wire did not help loading. They claim a
maximum loading ratio of 0.84. Shikano et al. (123) have loaded palladium foils covered on
one side with gold or silver. They showed that the electrical resistance increases with loading,
then decreases, as predicted by the resistance ratio versus loading curve (124). Li et al. (125)
have shown that they could load deuterium in palladium wires up to a ratio of 0.78. They
measured the palladium grain size, which were 100 to 200 μm. Huang et al. (126) showed that
the resistivity anomaly observed on the resistance ratio to loading curve was due to the alpha-
beta phase transition. Del Giudice et al. (127) have developed a technique capable of operating
at low temperature. They used a wire one meter long with a cross section $2 \times 50 \mu m^2$, made by
sputtering palladium. Scaramuzzi (128) has shown with a system similar to the previous one
that he could observe a resistance ratio between pure palladium and palladium loaded with
deuterium of 2.64 at 30K. This value is much larger than the one usually measured at room
temperature: 2.0 for a loading ratio of 0.74. Celani et al. (44) have shown that by depositing
nano-particles of palladium and other elements of nanometric size on the surface of a palladium
wire, they obtained ultra short loading times (of the order of 10 s with a Pd wire of 50 μm in
diameter. Most of the work on gas loading has been performed with palladium. However,
Shrikhande et al. (129) have tried to measure the resistivity of titanium versus loading, but the
results were irreproducible. Arata and Zhang (130) showed that in palladium loadings up to
three deuterium atoms per palladium could be obtained in nano-particles of palladium
embedded in ZrO$_2$.

10 – Transmutations
Transmutations are a by-product of Cold Fusion, they have been observed on electrodes after
electrolysis, but they remain doubtful because of potential contaminations by impurities always
present in the electrolyte. In gas phase, there is not such a danger, and the results are more reliable. Kong et al. (131) and Qiao et al. (132) have detected several new metals on palladium in a deuterium atmosphere. Iwamura et al. (133-135) have pioneered this field. In their first paper (133), they use a palladium foil as a substrate on which multi-layers of thin films of palladium and low work function materials such as CaO, TiC, Y$_2$O$_3$ ... are deposited. They flow deuterium gas through the sample by placing a pressure of deuterium gas on one side, and pumping on the other side. They analyze in situ, without bringing the sample to atmosphere by Photo-Electron Spectroscopy the surface composition, and by Secondary Ion Mass Spectroscopy their isotopic distribution. In their first experiments they show production of sulfur with an anomalous isotopic distribution: S-33/S-32 is an order of magnitude larger than the natural distribution. They also observed the presence of silicon and magnesium. In another experiment, they doped the surface of the multilayer with lithium. They observed a decrease in lithium with time, and an increase of aluminum. Iwamura et al. (134) added Cs and Sr on the surface of their sample and observed the transmutation of Cs into Pr and Sr into Mo. The interesting aspect here is that the isotopic distribution of the molybdenum is not natural. This is a direct proof of the non-natural origin of the molybdenum. These transmutations correspond to adding four deuterons to the initial nuclei. Later Iwamura et al. (135) have shown than in other cases (Ba deposit), the transmutation consists in the addition of six deuterons to the initial nucleus. Several groups have reproduced Iwamura’s work. In a similar experiment, Higashiyama et al. (136) have shown that the praseodymium formation is a function of the flow rate of deuterium through the sample. Minari et al. (137), Yamada et al. (138,139) and Kitamura et al. (140) have also observed transmutation. Castellano et al. (141) have shown that new elements are found on a deuterated palladium foil irradiated with a laser beam. Di Giulio et al. (142) have deposited thin films of palladium on silicon wafers. After laser irradiation they observe several transmutations products. Wei et al. (143) observed new elements (Gd, Tb, Nd) after diffusion of deuterium through a palladium foil.

11 – Nickel-Hydrogen system

Most of the work in gas loading has been made with the palladium-deuterium system. However, the nickel-hydrogen system is also of great interest. Sankaranarayanan et al. (92) reported tritium production with hydrogen in nickel wires. A team at the Siena University in Italy has performed some very promising work. Focardi et al. (144-150) have studied the nickel-hydrogen system. They have repeatedly produced excess heat. Battaglia et al. (151) have shown the production of neutrons, and Campari et al. (152,153) have shown the production of heat, gamma rays and neutrons. Focardi et al. (154) show the production of heat, particle emission and gamma rays. Campari et al (155) have observed anomalous distribution of elements on a nickel alloy during experiments with hydrogen. All the work mentioned above is the product of a single team. It is interesting to know that Cammarota et al. (156) have reproduced the excess heat measurements. However, E. Cerron-Zeballos et al. (157) could not succeed. A different approach has been developed by Mastromatteo (158). He used a silicon based hydrogen source to load a nickel film in a small device. He observed anomalous melting of the nickel film.
12 - Conclusion

Gas loading is a very promising technique. During the past twenty years, it has been shown that all the ingredients of Cold Fusion have been found with this method: excess heat, neutrons, tritium, gamma rays and helium. However most of the work has been done with palladium and deuterium. It is interesting to note that for future applications the nickel-hydrogen system is very promising, and deserve a lot more attention.

References

38. Y. Arata and Y. Zhang, in Tenth International Conference on Cold Fusion, Cambridge, USA, 2003, pp. 139.
82. A. S. Roussetski, in Sixth International Conference on Cold Fusion, Lake Toya, Hokkaido, Japan, 1996, pp. 345.
104. E. Botta, T. Bressani, D. Calvo, C. Fanara and F. Iazzi, in Sixth International Conference on Cold Fusion, Lake Toya, Hokkaido, Japan, 1996, pp. 29.
123. K. Shikano, H. Shinojima, and H. Kanbe, in Fifth International Conference on Cold Fusion, Monaco, 1995, pp. 251.
126. G. S. Huang and X. Z. Li in Sixth International Conference on Cold Fusion, Hokkaido, Japan. 1996, pp. 198.
133. Y. Iwamura, T. Itoh and M. Sakano, in Eighth International Conference on Cold Fusion, Lerici, Italy, 2000, pp. 141
158. U. Mastromatteo, Eighth International Conference on Cold Fusion, Vancouver, Canada, 2000, pp. 81.