

Arata and Zhang's Cold Fusion: Excess Heat and Helium Production

A Review by Mike Carrell

Introduction

Outstanding among the many confirmations of the pioneering findings of Fleischmann and Pons (F&P) is the work of Prof. Yoshiaki Arata and Prof. Yue-Chang Zhang, (A&Z) both of Osaka University in Japan. A special 56-page issue of the *Journal of the High Temperature Society* (HTS) of Japan¹ was devoted exclusively to a paper summarizing their work. **This work unambiguously links the production of substantial, robustly generated excess heat with the production of helium in a system involving electrolytic loading of deuterium into palladium metal.**

My purpose in this essay is to present this work to the readers of *IE* in a readable form, answering a number of concerns that have been raised in critiques in certain Internet groups. The 1997 A&Z HTS paper is a summary, omitting details covered in their earlier papers. In this essay I blend information from several papers into a coherent narrative. In a few cases, I offer hypotheses to explain certain features of the data not explained by A&Z, which I have clearly indicated. Some of the figures in the text have been traced from scanned images available from several papers. The tracing clarifies the images and in some cases I have added explanatory annotations. In all cases, the figures are a faithful representation of the data in the A&Z papers.

About Professor Arata

Osaka University in Japan has named a building after him—such is his fame as a top-ranking scientist. His address is Arata Hall and a booklet issued by Osaka University listing his accomplishments requires 40 pages. He was awarded the Emperor's Medal, the only physicist so honored in Japan. Arata was involved in Japan's experimentation with high-energy plasma fusion (hot fusion) as early as 1958. The collaboration with Ms. Zhang (Professor in the Welding Institute) began then, with early investigation of fusion in solid bodies. With the F&P announcement in early 1989, the focus of their work shifted to electrolytic cells, and they were able to announce positive results in 1990. In Japan, it is acceptable

and not unusual to work on both hot and cold fusion processes.

Why Aren't You Dead?

When Fleischmann and Pons announced that they had produced abnormal "excess" heat from an electrolytic cell using heavy water and a palladium cathode, and that they thought that a nuclear fusion reaction might be involved, the critic's question became "Why aren't you dead?" If the *known* hot fusion reactions were producing the "excess" heat claimed, these reactions, it was *said*, would also produce high and perhaps lethal doses of penetrating radiation. (Whether it would actually have been *lethal* is a matter for exact calculation of dosage, but certainly the radiation would have been significant and easily detectable.) Clearly Fleischmann and Pons and their graduate students were very much alive and well. What, then, was the source of the heat? Therein lay the essence of the emerging cold fusion controversy.

The Rutherford Fusion Reaction

The US and other governments have invested billions of dollars over many decades in projects to produce controlled fusion of hydrogen isotopes, which yields high energy output per fusion reaction but which also requires high energy input. Hot fusion requires forcing isotopes of hydrogen together, which is resisted by the mutual repulsion of the positively charged nuclei, until a critical distance between them is reached. Then the *strong nuclear force* takes over and the nuclei fuse, only to break apart in sec-

ondary reactions—releasing energy from mass conversion after the equation $E = mc^2$, where m is the mass lost in the reaction. The fusion conditions are very difficult to produce, requiring a star, a thermonuclear explosion ("H-bomb"), or very expensive magnetic or laser machines, that is, until the F&P experiment done in an electrochemical cell. There are several well studied fusion reactions. One of these, the Rutherford reaction, is the one F&P anticipated. In Table 1, it is compared with the initial results of F&P and those of A&Z.

Table 1 indicates the foundation of the "Cold Fusion" controversy. Governments had invested billions in a reaction involving tritium and deuterium, because the deuterium-deuterium reaction is much harder to do. In Tokamaks, brief bursts of power nearly approaching the operating power have been seen, but over the long term the energy yield is less than the energy required to run the system. While the fusion reactions in Tokamaks do not themselves produce radioactive waste, the high energy neutron radiation which penetrates the reactor walls to produce useful power also makes them radioactive. (Moreover, the neutrons generate the needed tritium fuel for the reactor (bootstrap fashion) by neutron bombardment of molten lithium metal surrounding the chamber.)

Now F&P claimed safe over-unity heat production from a fusion reaction that did not have the usual radiation signatures. Attempts to duplicate F&P results usually produced feeble, if any results. There were also several positive results that were deemed "negative" by critics bent on disproving F&P. Critics claimed

Reaction Product	Rutherford	Fleischmann & Pons, 1989	Arata & Zhang, 1989-97
Helium 3 (³ He)	50 %	Note 1	Yes
Tritium (T or ³ H)	50%	Note 1	No
Helium 4 (⁴ He)	1 : 10 million	Note 1	Yes ⁴ He/ ³ He = 4
Gamma Radiation	23.8 MeV energy	Note 1	Note 2
Neutron Radiation	2.45 MeV energy	Low level claimed, retracted.	Note 2
Energy Out/In > 1	Theoretically yes, not achieved in practice.	Initially small, elusive Later 100s of Megajoules	Robust, 200+ Megajoules per cc of active material

Note 1: F&P did not test for ³He or ⁴He; they did test for tritium (T) and found it. (Many other investigators of the F&P cell found higher levels of T later). Helium has been found by several other investigators, including A&Z, but not in the proportions expected from the Rutherford and other known nuclear reactions. F&P did not detect gamma radiation.

Note 2: A&Z did not report gamma and neutron radiation, nor the search for them, in the papers I have seen.

that the measured heat was from obscure chemical reactions, or energy storage, or the result of faulty measurements. F&P did not emphasize how difficult it is to produce the effect. In retrospect from 1998, it is possible to see what the problem was, now that many successes have been confirmed. **Among these is the work of A&Z, who have demonstrated in a vivid way the essential correctness of F&P's vision. They show strong evidence of a nuclear fusion reaction that does not fit the conventional rules.**

The Cathode Problem

Metallic palladium is a three dimensional lattice of palladium atoms spaced evenly in a face-centered cubic array (atoms at each corner of a cube and the center of each face). Within this lattice there is space for hydrogen or deuterium nuclei (deuterons, D⁺). There are several ways to load deuterons into palladium, such as: a) gas pressure, b) ultrasonic cavitation in heavy water, and c) electrolysis —the method chosen by F&P.

Deuterium is readily absorbed in two phases. From the gas phase D₂ gas is dissociated by catalytic action at the surface and deuterons enter the palladium lattice. In a F&P cell, D₂O is dissociated by electrolysis at the cathode; part of the deuterons combine to form D₂ gas which escapes as bubbles, another part diffuses into the cathode metal lattice.

Initially, in the alpha phase, the deuteron concentration is low. The deuterons are essentially in solution, and free to diffuse or migrate. As more deuterons are pumped into the lattice, they interact with each other and the lattice and find positions of equilibrium among the palladium atoms. This is the beta phase. At this point the cathode swells and cracks may appear from internal stresses, allowing the deuterons to leak out and form D₂ gas.

In the solid cathodes used in traditional F&P cells, the electrolyte surrounds the cathode and deuterons diffuse in from all directions. Voids in the cathode metal are sinks where deuterons can accumulate as D₂ gas under pressure, delaying the transition to the beta phase. *A&Z turned this to an advantage*, which we will see below.

F&P reasoned that electrolysis could be used to generate the equivalent of high gas pressure to force still more deuterium into the palladium lattice, and that under such pressure and proximity of deuterium atoms, fusion reactions might occur. From a 1998 perspective, many experi-

ments indicate that the loading must be such that 85% or more of the possible deuteron sites are occupied for the reactions to begin.

Loading and maintaining palladium at this level is quite difficult. Not all samples of palladium will load well, and even those which load may not show the F&P effect. Best results are reported for palladium from a single supplier, ordered from a single production lot of metal (perhaps containing special beneficial impurities). Yet researchers report that those cathodes which do work will work in many cells.

F&P did not emphasize these difficulties. Many researchers thought the experiment looked easy, reached for a handy lump of palladium and some heavy water, and tried the experiment. A few succeeded. Many ran the loading phase of the experiment for weeks or months without seeing any result, or feeble, transitory effects. Critics claimed that obscure chemical reactions were responsible for any heat seen, or that the energy put in the cell during the long loading phase was merely being returned in a short burst, like a charged battery.

To this day, there is no accepted theoretical guide to the conditions which produce the F&P effect. There are recipes which work for various experimenters, and it is clear that electrolysis is only one way of producing the necessary loading, or concentration of deuterons within the Pd lattice.

As early as 1958, A&Z had been investigating ways of producing fusion in solids, and had worked with shells and cylinders of palladium and with colloidal (finely divided) palladium, or palladium black (Pd-black). With the F&P announcement in 1989, they decided that the reaction occurs near the surface, not deep within the bulk of the cathode. This led to their development of a "double-structure" (DS) cathode, which proved very successful.

The Arata & Zhang Double Structure (DS) Cathode

The DS cathode is machined from a solid rod of palladium, forming a capsule with a cavity in one end which is sealed with a welded plug. The cavity is packed with Pd-black under vacuum conditions, so that it is substantially filled with the Pd-black in contact with all the walls and filling the cavity.

In some experiments, a pressure gauge was attached to the capsule to measure internal gas pressures. Thermocouples were also used to measure internal and surface temperatures in other experi-

ments. In the critical energy runs discussed below, the capsule was sealed without attachments or probes except the electrical connection for its function as the cathode in an electrolytic cell.

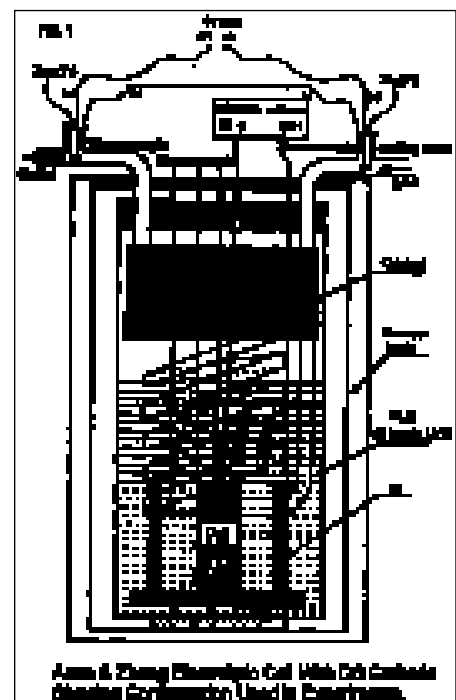
A&Z avoided many of the problems of solid cathodes by using Pd-black. With particles as small as 40 nanometers across (about a hundred atoms), it was practically all surface and could hardly be broken into smaller particles by the stresses of loading deuterium gas. I discuss the processes within the DS cathode a bit further on.

Robust Performance of the DS Cathode

A&Z were working with the DS cathode structure at the time of the F&P announcement. From 1990-92 they ran many trial-and-error experiments. In 1993 they gave a preliminary report to ICCF-4. From February 1993 to May 1994 they continually experimented with excess heat and reproducibility, which included cycling the cell on and off every few days. The cell was idled for a year, then restarted for a 3000-hour run (at the time of a report dated 23 Dec. 1994). This 1994 report shows excess heat vs. time curves before and after the year-long shut-off; the performance is substantially the same.

The A&Z Test Cell

Fig 1 shows a schematic diagram, reproduced from an A&Z illustrations. It shows the principal features of their cell in their functional relationship as used in experiments.



Closed Cell. The cell itself is closed. The electrolysis releases oxygen and deuterium gases, which carry heat energy out of the pool of electrolyte and would result in a potentially explosive buildup of gas pressure if it were not for the recombiner catalyst.

Recombiner. Above the pool of electrolyte is a porous body containing a catalyst, usually platinum, which induces the oxygen and deuterium gases to recombine, forming heavy water (deuterium oxide), which drips back into the electrolyte solution. This returning fluid also returns the heat energy removed as gases.

Insulating Jacket: The cell is contained within an insulating jacket which is often a vacuum bottle, or Dewar flask, but can be of any highly insulating construction. Heat can escape from the cell by many subtle paths, even "leaking" along electrical conductors and supports. Calibration is necessary, which A&Z did.

Cooling Coil. The cell is cooled by water flowing through a helical glass coil, substantially submerged in the electrolyte pool. The inlet and outlet temperatures of the water are measured by thermocouples attached to recording equipment. The flow rate is measured by an instrument not shown, also connected to recording equipment. Knowing the flow rate and the temperature differential between inlet and outlet, the heat produced within the cell is easily calculated.

Anode. The anode is a cylinder of platinum, about the length of the cathode and the diameter of the cooling coil, located just below the cooling coil, and connected to an external power supply by an attached wire which exits through the cell closure.

Cathode. The cathode is a capsule placed at the bottom center of the cell, connected to the external power supply by a wire which exits through the cell closure.

Calorimetry Considerations

It happens that calorimetry, the measurement of heat, is something that looks simple but is a maze of subtle traps for the unwary. It is closely related to thermometry, the measurement of temperature, which is only slightly less hazardous for the unwary. Yet it is the production of "excess" heat which is the key signature of the new energy reactions. It takes a certain amount of energy to operate an electrolytic cell, which will produce heat, release gases, and produce other chemical reactions. In the F&P effect, after all ordinary sources of heat and chemical energy are accounted for, an "excess"

heat release is found. At first, the "excess" was small and critics cited various kinds of measurement errors as the source of apparent "excess." Later experiments showed robust, powerful heat release, in the range of hundreds of watts, on a continuing basis. A full discussion of calorimetry is beyond the scope of this article.

Calorimetry of the A&Z Cell

Two basic approaches to calorimetry are *static* and *flow calorimetry*. In static calorimetry, the cell is isolated within an insulating jacket, all fluids are retained, and the temperature is maintained constant. Usually, the cell is maintained at a constant elevated temperature by a calibrated electric heater. Heat produced by a reaction inside the cell reduces the power necessary to the heater to maintain the constant temperature. This method requires careful calibration.

Flow calorimetry, chosen by A&Z, removes heat by means of water flowing through a cooling coil, as seen in Fig 1. It is only necessary to measure the inlet and outlet water temperatures, and know the flow rate, to calculate the heat produced. The method is self-calibrating.

Some cell configurations used by other researchers have been criticized for measuring temperatures at single points, which may not be representative of the average temperature, and for inadequate mixing of liquid. A&Z avoid these problems by the features shown. The central, lower location of the cathode capsule encourages a general circulation of the fluid, rising in a warmed plume from the central cathode, and sinking around the outer periphery, enclosing the cooling coils. Gas bubbling from the anode and cathode further mix the fluid.

Inlet and outlet temperatures are measured at single points, but these are within the bore of a tube carrying water which circulates in a spiral through the electrolyte pool, mixing and exposing all sides to the surrounding fluid before exiting. Thus while a single point temperature measurement of exit temperature is made, it is representative of the average temperature of the whole body of electrolyte.

Calibration of the A&Z Cell

Calibration is accomplished by substituting a platinum cathode of the same size and shape as the palladium cathode and running the electrolysis reaction at representative power levels. Platinum does not produce the F&P "excess heat," but all other features of the cell and its support system are the same. Fig 2 shows

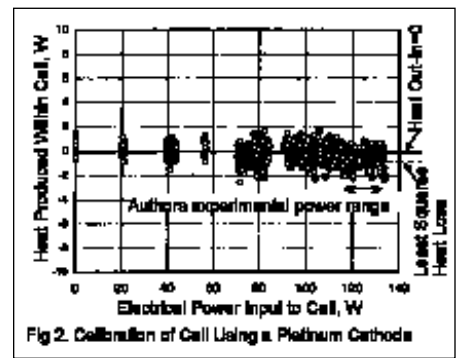


Fig 2. Calibration of Cell Using a Platinum Cathode

the results of such a calibration, reproduced from A & Z with labels modified for clarity. Fig 2 shows heat produced within a cell using a platinum cathode of the same size and shape as the palladium DS cathode. By applying least squares statistical analysis to the data points, A&Z show that the cell loses about 0.7 watt of power when operated at its normal range of 120-135 watts input. There is a scattering of data points within a range of +/- 2 watts peak-to-peak, but mostly clustered between +/- 1 watt. These numbers are important in evaluating data presented later.

It will be shown that this variability does not obscure the measurement of very large energy release from the A&Z cell, but some critics have seen this variability as an indication of experimental weakness.

The Energy Measurement

Fig 3 gives a plot of excess power from an A&Z cell with a palladium DS cathode capsule filled with 3 grams of Pd-black. In this graph, the horizontal 'zero' line represents the heat equivalent of the electrical input power to the cell. All points above this line represent measured "excess" power produced by the processes in the DS cathode. The run extends for about 4800 hours. A&Z state the released energy (integrated power over time) to be in the range of 200-500 me-gajoules/cm³ of active material (the Pd-black). This is far, far beyond any possible chemical reaction. It is far beyond any government hot fusion program, where the energy released has never exceeded the energy necessary to run the experiment. Only nuclear reactions are known to produce this kind of energy density, but the expected radiation signatures of the Rutherford reaction are missing and Arata and Zhang remain in good health. The data looks noisy and variable, which has led some critics to assume that the measurements are not those of a real operating process but consist entirely of artifacts of a poorly constructed experiment. What is truly important about Fig 3 is that the plotted energy is in all cases

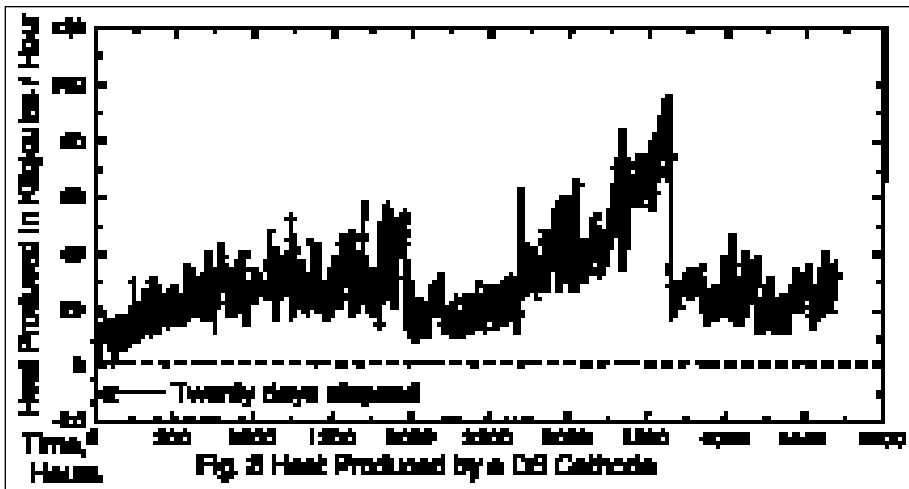


Fig. 3 Heat Produced by a DS Cathode

clearly above the reference line. Whether variable or not, all points represent energy released from the DS cathode. Summed over the 4800 hour duration of the experiment, hundreds of megajoules per cubic centimeter of active material were generated. This is to be compared with the slight (0.7 watt) average heat loss from the cell during the calibration runs. Because of the importance of the energy measurement, this will be discussed in some detail in a later section.

The Mass Spectrometry Measurement

The energy release indicates a nuclear reaction, but a nuclear reaction should leave tracks, a signature. The expected signature of the Rutherford D-D fusion reaction was given in Table 1. The penetrating radiation is not seen, but what about the "ash," helium isotopes ^3He and ^4He and ^3H (tritium)? Other investigators have claimed to see these products, but the method of analysis is always challenged by critics. Traces of helium are in the environment and are very difficult to exclude.

The standard method of detecting is by a quadrupole mass spectrometer (QMS). Mass spectrometry in general proceeds by vaporizing a sample, ionizing it by an electric field, and then moving the ionized particles by a combination of magnetic and electrostatic fields such that they take curved paths whose radius depends on the mass, and counting the number of particles that fall on a detector for each setting of the controlling fields. Knowing the conditions, the quantity of particles of each mass can be measured. A quadrupole mass spectrometer is simply one in which four magnetic poles are used in a very compact design. A&Z were familiar with well-known drawbacks of conventional QMS, so they designed a special system which

avoids them. It is shown schematically in Fig 4.

General Construction: Stainless steel, with metallic gaskets and gate valves, standard practice in high vacuum work. The valves can seal off each component of the system as needed. QMS(M3) and QMS (M4): Two quadrupole mass spectrometers are provided, one primarily tuned to look for Mass 3, which includes compounds of D and H (including ^3H , or tritium), and ^3He . The other is tuned to Mass 4 (which includes deuterium gas D_2) and the normal isotope ^4He . Both QMS can scan for other masses as well.

Standard Mix Gas Vessel: This contains a standard gas used to calibrate the mass spectrometers.

Molecular Pump: This is essentially a very high speed turbofan made of stainless steel which sweeps any molecules entering it back towards a positive displacement pump which removes the concentrated gas from the system.

Getter Pump: A getter is like "molecular flypaper"; surfaces coated with a small family of very reactive metals will capture gas molecules of all types except helium, which is chemically inert. (A generous getter coating on the inside of television picture tubes maintains the necessary high vacuum over many years, even though organic binders are used with the phosphors on the faceplate which produce the light). Getters will produce harder vacuums than molecular pumps. Usually the getter is initially in the form of oxides in a small cup. When most gas has been removed by other pumps, the oxides are heated, evaporating them, coating the active metal on a passive surface while the oxygen is pumped out.

Sample Chamber: Pd-black removed from an energy run such as shown in Fig 3 is placed in this chamber, where it can be heated to 1300 C.

QMS Operation

The basic operational sequence is as follows (variations were used in special cases):

- 1) Place the sample in the sample chamber and seal the valve to the outside world.
- 2) Open all internal valves and start the turbomolecular pump and its backup pump.
- 3) Pump for 5-15 hours to a pressure less than 10^{-8} Torr.
- 4) Activate the getter pump.
- 5) Close the pump valve and stop pump.
- 6) Heat the sample in step-wise increments to 1300°C
- 7) Scan QMS repetitively over the mass ranges of interest, Mass 3 and Mass 4

What happens in this sequence is that the Pd-black particles are initially coated with various pollutants from exposure to the laboratory environment before and after the heat generation in the DS cathode noted above. These pollutants are driven off by the progressive heating. Since they are chemically active, they will be captured by the getter pump. Then, as the heat increases, deuterium and helium isotopes (created by the nuclear reactions) diffuse out of the Pd-black particles and are measured by the two QMS units.

Deuterium is an isotope of hydrogen and shares its chemical activity; it is captured by the getter and removed from the "Closed QMS" system in a period of minutes. Helium is chemically inert. It is not absorbed by the getter and remains in the system. Deuterium gas (D_2) and ^4He have nearly equal masses, but the QMS is able to distinguish between them. While D_2 is present, it serves as a calibration marker. It is important to understand that helium is nearly insoluble in palladium; therefore, any helium appearing in the "Closed QMS" system must come from within the Pd-black, and so must have been created from deuteri-

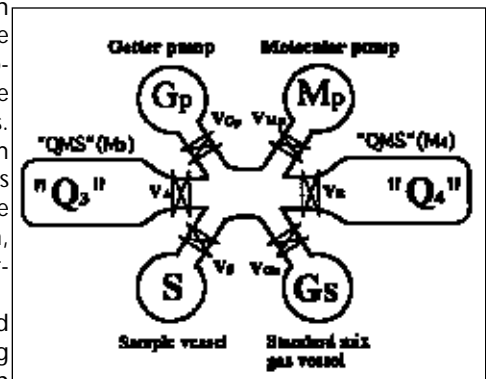


Fig. 4 Schematic Configuration of "Closed QMS" System

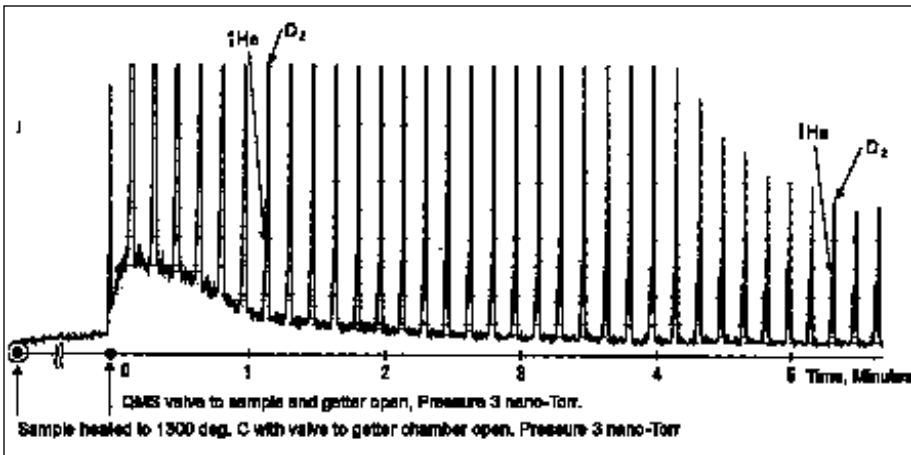


Fig. 5 QMS Scan (Mass 4 Range) of Gases From Heated, Heavily Deuterated Pd-black

um by a nuclear process.

Fig 5 shows a graph produced by the QMS. It is set to repetitively scan over a narrow range centered on a mass of 4. Any particles whose mass is near four times the mass of a hydrogen ion will register, the vertical height being the intensity, or population at that mass number. The sample is heated for about a minute in the evacuated system, with only the getter and sample chambers open. The valves into the QMS are then opened. At this point surface contaminants evaporated from the sample surge into the QMS, producing the hump. Yet, very strong peaks from the D_2 desorbed from the sample are seen. These give an immediate calibration to the QMS display. Very small peaks at the left edge of the D_2 peaks are initially seen, which is where 4He is expected. By the fifth minute the getter has absorbed enough of the D_2 that its signal is reducing and the 4He signal remains, even growing

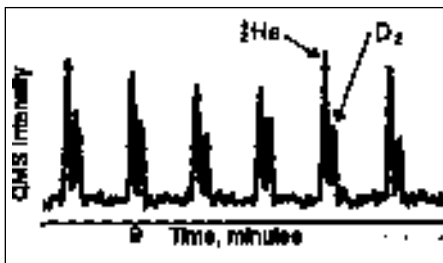


Fig. 6. D_2 - 4He Mass Spectrum After 9 Minutes

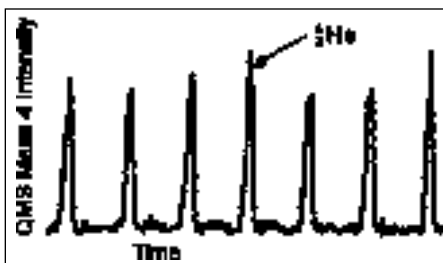


Fig. 7 QMS Mass 4 Spectrum After 67 Minutes. System at Room Temperature

stronger as it continues to come out of the sample but is not absorbed by the getter. By the ninth minute the helium signal dominates, as seen in Fig.6.

The sample heater is then turned off, and the system allowed to cool to room temperature, with the getter chamber still open.

At 67 minutes, the Mass 4 spectrum, shown in Fig 7, now shows only one peak, that of 4He .

In Fig. 8, the second QMS is tuned to the Mass 3 range, and at 67 minutes shows 3He with a marker of the compound DH, whose mass is nearly 3. The DH is mostly absorbed by the getter at this point, since it is chemically active.

In these graphs we see clear evidence of two isotopes of helium being driven from within the Pd-black particles. It is important to understand that helium is virtually insoluble in palladium. Helium which appears and remains in a sealed system as all chemically active gases are absorbed by the getter must have come from within the Pd-black sample, created there by the reactions which also generated the excess heat. The graphs shown here are for a particular sample but are representative of about a dozen samples whose mass spectra are shown in the 1997 HTS paper.

Anomalous Heat and Ash: A New Nuclear Reaction

It is now time to review. A&Z built an

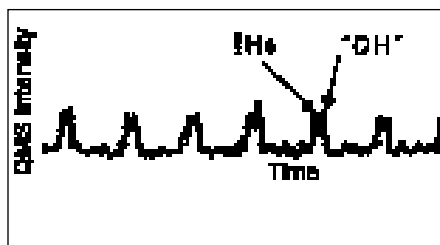


Fig. 8 Mass 3 Spectrum After 67 Minutes. System at Room Temperature

electrolytic cell using heavy water with a cathode consisting of a sealed palladium capsule containing a few grams of Pd-black. Over a period of 4800 hours this cathode generated several hundred megajoules of heat energy above and beyond the energy necessary to run the cell. At the end of the run, the Pd-black was removed and heated in a sealed mass spectrometer which showed that 4He and 3He had been created within the Pd-black. 3He is rare in nature, and neither isotope of helium readily dissolves in metals.

Helium and heat are two signatures of the deuterium fusion reaction. Table 1 lists the reaction products of the expected Rutherford DD reaction. A&Z's reaction does not match the signature of the Rutherford reaction, yet the heat energy release places it completely outside of any chemistry. Further, A&Z found no tritium in tests outlined below.

These results meet the critical demand for coupling of heat and nuclear ash in one experimental context. It is good news that the heat energy comes without intense radiation or radioactive ash. It is bad news for conventional physics because the experimental results do not match any known DD reactions. A&Z propose that in a palladium lattice saturated with deuterium, intense local vibrations ('lattice quakes') and microplasmas may occur which enable novel fusion reactions to take place.

Details, Details, Details

Confidence in A&Z's results comes from careful study of their work, which began before the F&P announcement in 1989. We will now turn to details which aid in understanding the thoroughness of their work.

Calorimetry Questions

There are a number of questions to be answered about the calorimetric data shown in Fig 3.

- 1) What causes the short term variation in the heat generated, that looks like noise?
- 2) What are the operating parameters of the cell: voltage, power, temperature differential, coolant flow?
- 3) Are the temperature measurements representative of the significant temperatures in the cell?
- 4) What causes the large scale variations in heat generated; the burst at 1800 hours, the drop at 3700 hours?
- 5) The cell input power is in the range of a hundred watts, the excess is a few watts. How can we be confident that the

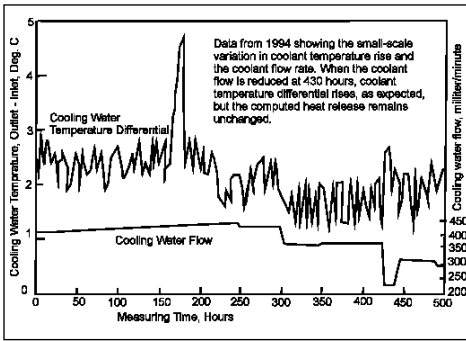


Fig. 9 Coolant Temperature Differential and Flow Rate for a DS Cathode

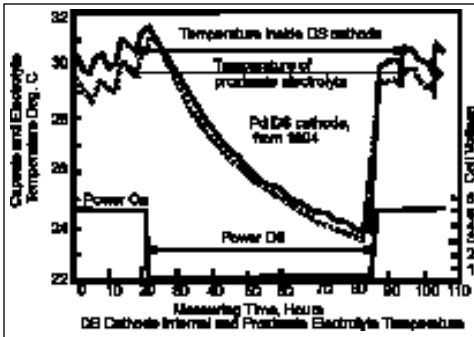


Fig. 10 Capsule Interior and Proximate Electrolyte Temperatures and Cell Voltage

results are not artifacts of a poorly constructed experiment?

6) Does the cell respond to changes in external parameters?

Short Term Variation and Operating Parameters

The Y-axis of Fig 3 gives energy in kilojoules/hour (KJ/hr), where one watt-hour is 3.6 KJ. The graph is normalized such that zero on the Y-axis is the input electrical energy for every plotted point; the distance above the zero line is "excess energy." The X-axis is process time, in hours. The experiment ran for about 4800 hours. The data is very crowded, but plots of other experiments indicate that data is recorded at least hourly, if not more frequently in some cases. Automatic data logging equipment was used.

The short term variations are present whenever the cell is powered. A&Z offer no explanation for this, but a hypothesis which fits the facts is as follows:..

As I mentioned above, heat energy is carried in the evolved gases into the recombiner at the top of the cell. The catalyst causes heavy water to form, releasing energy as heat. This water is not immediately returned to the electrolyte pool, but may be retained for a time (as atmospheric moisture may accumulate in a cloud) before "raining" back into the electrolyte, return-

ing the heat energy lost. This process may be quasi-cyclic, producing cyclic variations in the temperature differential measured and the apparent instantaneous heat output of the cell. Since the interval between plotted points is probably not that of this quasi-cyclic process, irregularity in the plotted values can be expected. This is an artifact of the electrolysis process both in the calibration with the platinum cathode and in operation with a palladium cathode. It disappears whenever the electrolysis power is turned off, even though heat production may continue in the case of the DS cathodes.

Support for this hypothesis is in Fig. 10, which shows temperature measurements made in the capsule and in the proximate electrolyte. There is a cyclic variation of about 1° C peak-to-peak, with a period of about six minutes. The plotted oscillations are so nearly in phase that it is not possible to determine if the variations originate within the capsule or in the proximate electrolyte in the cell. Only a few millimeters of palladium and some electrolyte separate the two probes. It is not clear what processes within the capsule, or at the capsule-electrolyte interface, could cause such cyclic variations. Variations according the above hypothesis, producing changes in the electrolyte temperature, might be conducted to the capsule interior within the time scale shown.

There may be other processes at work. All measurements are not available. The electrolyte pool is well mixed by convection currents and bubbling, which should not show cyclic variations of the type seen.

Fig 9, from a 1994 paper, gives us more information. Here the short term variation is plotted in terms of the differential temperature between inlet and outlet cooling water. The variations fit within an envelope ~0.8° C. We also find a value for the range of water flow, a few hundred milliliters per minute. We also see that when the flow is cut, the temperature differential rises, as would be expected. The computed power yield is unchanged. The measurement changes in response to a change of parameters and is thus not a self-generated artifact.

One critic speculated that boiling might occur in the cell, creating a froth that might cause irregular measurements. Fig. 10 shows data from another test in 1994 in which temperature inside the DS and in the electrolyte just beside the capsule were measured. The absolute temperature in the range of 28-30° C, well below boiling, and the temperature inside the capsule is only a few degrees higher than the surrounding electrolyte. A cyclic variation in temperature is seen

while the cell is operating, which may be due to the "raining" of condensate from the recombiner or to another, unidentified cause, as discussed above.

Note that the variation disappears when electrolytic power is shut off, with an exponential decline in heat production over a period of tens of hours, as one would expect from the depletion of deuterium fuel in the CF reaction.

Representative Measurements

A criticism made of some CF experiments is that thermocouples used measure temperature at only one, rather small, point, which may not be representative of the whole cell and by being at a fortuitous 'hot spot' may give over-optimistic results. I should mention that this criticism is not valid for other, more recent CF experiments, and has no bearing on the A&Z cell.

Fig. 1 shows the capsule centered at the bottom of the cell. Electrolyte warmed by the internal heat of the capsule rises in a plume to the top of the pool and returns down the coil in which the cooling water circulates. In addition, gases bubbling from the cathode and anode will further mix the liquid.

Cooling water enters the cell by a coiled tubing which reaches down to the top of the anode. Inlet and outlet temperatures are measured by two pairs of thermocouples. One pair provides independent measurements of the inlet and outlet temperatures. The other pair, connected in opposition, directly measure the temperature differential and thereby the heat produced within the cell. While these are indeed point measurements, the flow rate of ~300 ml/min indicated in Fig. 9 assures mixing of the water stream, which itself is exposed on all sides to the electrolyte and is thus representative of the heat produced within the cell.

Power Input and Output

Fig 11 is a cross-check on the calibration procedure noted above. Here is a new cathode, placed in a cell, and for 250 hours the output power closely follows the input power, with some very small variations seen in the output power. The Y-axis scale is in KJ/hr, where 1 KJ = 3600 Watt-seconds. Here one Joule = 4.2 (time, seconds) (volume of water) (temperature differential).

Then, suddenly, the reaction ignites and the output power is double the input power for hundreds of hours.

At one point the input power is turned off for 20 hours. The reaction does not immediately quit, but declines rapidly.

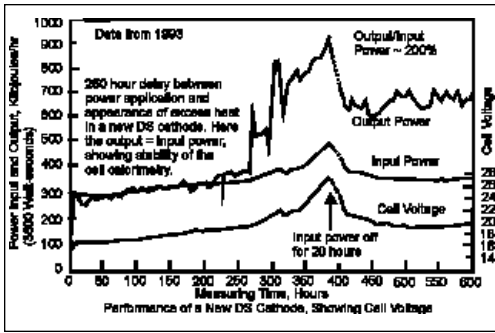


Fig. 11 Performance of a New DS Cathode, showing Cell Voltage

Once again, the heat production responds to a change in an external parameter, the power input to the electrolysis. The cell voltage is in the range of 15-30 volts.

Fig 12 shows the relationship between cell input electrical power and the heat generated, both given as watts, for the cell above. Note that before the reaction ignites, there is no excess heat. During the ignition period, there is a rapid rise in heat output without much change in input. After that, there is proportionality between the excess heat generated and the input electrical power. We will see later that this is consistent with other measurements which show that the rate that deuterium ions enter the Pd-black is proportional to the cell current.

Large Scale Variations

The large scale variations in excess heat shown in Fig. 3 were a puzzle during the early study of the A&Z work, and led one critic to characterize the data as "jumpy" or "noisy." Since the internal processes in the capsule are not well understood, it is possible that the large scale variations are the result of unknown factors — which do not invalidate the salient fact of energy yield. I suggest a plausible source of the short term variations above. The source of

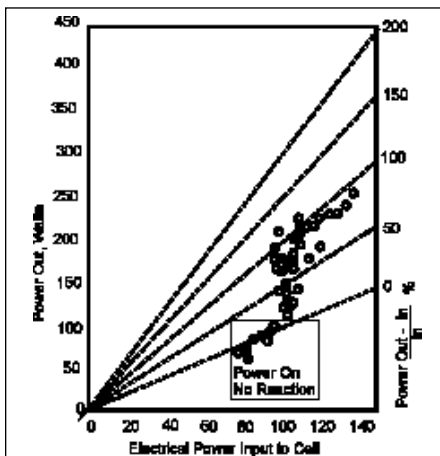


Fig. 12 Heat Power Out vs. Electrical Power Input for the DS Cathode of Fig. 11

the long term variations may be equally mundane: A&Z simply changed the cell input power. The graphs from papers dating back to 1994 show that A&Z changed the operating parameters during a run, apparently to explore their effect on the process. Several of these are shown in this article. There are also other plots of the type of Fig 12, showing a strong correlation between cell power and output power. We don't have an input power plot corresponding to Fig. 3, but it is credible that A&Z simply increased the cell input power gradually from ~2500 to ~3500 hours, then cut it back to a lower level. The drop at ~3300 hours is abrupt on the graph's time scale. Taking Fig. 10 as a model, the capsule temperature fell to that of the cooling water in ~60 hours, which would be a vertical line on a scale spanning 5000 hours.

Calorimetry Summary

We have answered the six questions posed above with evidence that the calorimetry is valid, that there are straightforward explanations for the small and large scale variations in the data, and have discovered reasonable parameters for the operating conditions. We can accept one major claim by A&Z: their DS cathode produces strong, sustained excess heat output of the order of megajoules per cc of active material.

The Search for Tritium

A QMS cannot directly distinguish tritium from Helium 3. Tritium is an isotope of hydrogen, with two neutrons and a proton in the nucleus. Helium 3 has two protons and one neutron. The two atoms have virtually identical masses. Tritium can form compounds with hydrogen, deuterium, and oxygen, but ³He forms no compounds, being a noble gas. If the Rutherford reaction is producing the excess heat, equal amounts of tritium (T) and ³He should be produced. The tritium can be expected to form compounds such as T₂, T₂H, TH₂, DT, D₂T, etc. at mass numbers 5,6,7. QMS runs were made with Pd-black which had generated heat in a test cell, and with Pd-black not loaded with deuterium. The QMS signals at the mass ranges 5,6,7 were the same for the deuterated and non-deuterated samples, indicating that no tritium had been produced by deuteration and heat generation in a test cell. The absence of tritium compounds is taken as further evidence that the Rutherford reaction is not involved.

The QMS graphs for this test are included in an appendix. They are complex and require careful study.

The Special Qualities of the DS Cathode

As noted above, the cathode properties are key to the performance of F&P cells. A&Z claim special advantages for their DS construction, indeed they were one of the first to show sustained, substantial heat production in a Pd-heavy water cell. The key ingredient is colloidal palladium, or Pd-black. It can be prepared by precipitation out of solution by a chemical reaction, or by electrically exploding a wire of palladium in an argon atmosphere (Argonide Corp.). The process used by A&Z is not known, but they claim a particle size range of 20 to 60 nm with an average of 40 nm. I have calculated for cubic particles 40 nm on a side, that the surface area of one gram of palladium is some 8300 square meters. This greatly enhances adsorption of deuterons.

Fig 13 shows pressure buildup in a stainless steel pressure vessel simulating a DS cathode, with a capillary leak of deuterium gas. If the vessel is empty, the pressure buildup follows a straightforward trend. If the vessel is filled with nickel powder, the pressure rises more rapidly at first because the powder reduces the effective volume of the vessel and does itself not absorb deuterium gas. The nickel curve remains above that for the empty vessel, since the effective volume is reduced. If the vessel is filled with palladium filings, the pressure initially follows the trend for nickel, but then drops as absorption takes over, yet soon rises as the surface of the particles is saturated. With Pd-black, the very large surface absorbs the gas completely until saturation occurs; then the pressure rises with the same trend as the empty vessel, but with a large displacement representing the absorbed gas. With a hollow palladium cathode, the diffusion of deuterons through the walls of the capsule is the equivalent of the leak in the previous experiment.

Fig. 14 shows the relationship between cathode current and cavity pressure for a

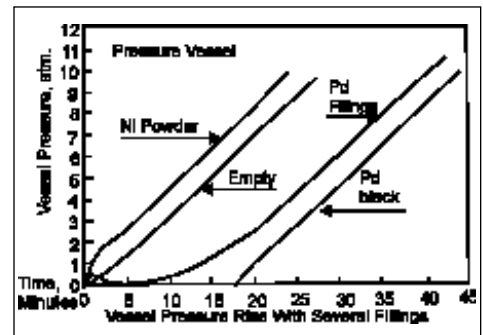


Fig. 13 Pressure Buildup in a Stainless Steel Pressure Vessel with Several Fillings

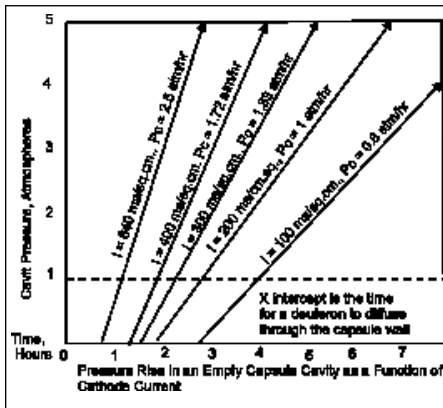


Fig. 14 Pressure Rise in an Empty DS Cathode as a Function of Cathode Current Density

particular cathode.

The first curve, for a cathode current density of 640 ma/sq.cm., is of special interest. There is an incubation time of ~0.8 hours from the start of cathode current to the start of pressure rise inside the capsule cavity. This is the time required for deuterons to diffuse through the capsule wall. The incubation time and rate of pressure buildup are both functions of the cathode current density.

When the cavity is filled, the pressure-time characteristic changes. In the usual solid cathode, deuterons migrate inward from all surfaces until the beta phase concentration is reached. The A&Z cathode has a cavity in its center, which makes a large difference. Deuterons reaching the inner surface face a potential barrier; they must find another deuteron and some electrons to form D_2 gas before they can leave the surface. Extra electrons are plentiful, for the capsule is a cathode and supplied with electrons by the external power source.

Spillover: Deuteron Surface Migration

Deuterons reaching the cavity wall can migrate over the surfaces of the packed Pd-black particles without surmounting the energy barrier to combine to produce D_2 gas. This migration is called *spillover*. For some time its existence was doubted in the chemical community. There now have been three international conferences on spillover, the last in 1993. The effect of spillover is seen for a DS cathode in Fig. 15

The empty cavity shows negligible incubation time. The current density for this experiment is not stated. With 2mm Pd beads, the pressure rise is similar to the case above with filings. As the particle size is reduced, the incubation period increases as the deuterons are absorbed in spillover migration across the surface

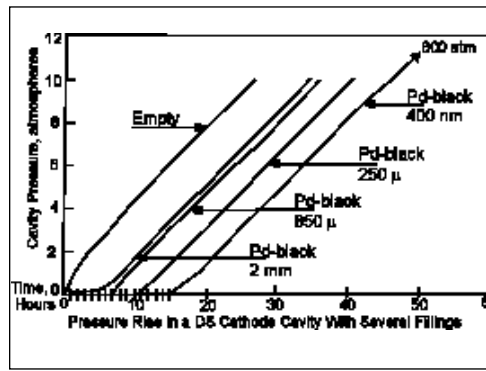


Fig. 15 Pressure in a DS Cathode with Several Fillings, Showing Spillover

of the Pd-black particles.

At the surface of the particles, energy is required for deuterons to combine into D_2 gas. The pressure rise will not begin until the surface is saturated with deuterons. Deuterons can diffuse into the particle lattice without surmounting the energy barrier necessary to form a gas.

Note that Fig. 11 showed a 250 hour period between starting a new cathode and ignition of the nuclear process. This is much greater than the incubation periods discussed here. A&Z do not discuss this. When power is switched off, the pressure in an empty cavity falls faster than in a cavity filled with Pd-black, as shown in Fig. 16.

Here a DS cathode was operated with a current density of 640 ma/sq. cm. until the internal pressure reached four atmospheres. The power was then shut off and the pressure allowed to decline. The incubation time and pressure rise/fall rates are shown in the figure. The effect of the Pd-black is seen again, both in increasing the rate of pressure rise, but more significantly in the reduced fall rate. When the power is off, the electrochemical potential driving deuterons into the cathode is gone, so deuterons within the cathode, including the cavity and the Pd-black, can diffuse outward. The substantial quantity absorbed will take longer to escape.

Pd-black Surface Contamination

A&Z mention that the Pd-black can be coated with surface contaminants from a few to ten atoms deep, comprising as much as ten percent of the particle volume. This remark most likely pertains to the 40 nm size, which is only a few hundred atoms across. This surface contamination is burned off in the early stages of the sample heating in the QMS.

Pd-black Melting

A&Z provide electron microscope images of 40 nm Pd-black before and after processing, which are shown as Figs. 17 & 18. Before processing, the particles have a distinct granular appearance. Afterward, the shapes are softened, as if by partial melting, with no evidence of internal cracks. Thus there is evidence for locally elevated temperatures, consistent with nuclear reactions.

Summary

The work of Arata and Zhang is one of several landmarks in the evolution of the Fleischmann & Pons discovery. From as early as 1990 they obtained robust, strong excess heat results with their DS cathode and went on to demonstrate clear evidence of nuclear reaction ash in the processed Pd-black powder. Criticisms of earlier F&P-class experiments have been answered or surmounted by the experimental procedure and apparatus.

Acknowledgments

Stimulus for careful study of the A&Z materials came from debates on the Internet Vortex-I forum with Kirk Shanahan and Rich Murray (who provided me with an English version of a 1994 A&Z paper). Thanks also to Akira Kawasaki for a painstaking translation of a Japanese version of the 1994 paper. Jean-Paul Biberian provided some valuable commentary on the spillover phenomenon. Peter Glück pointed me to a paper on spillover in the chemical literature.

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Papers:

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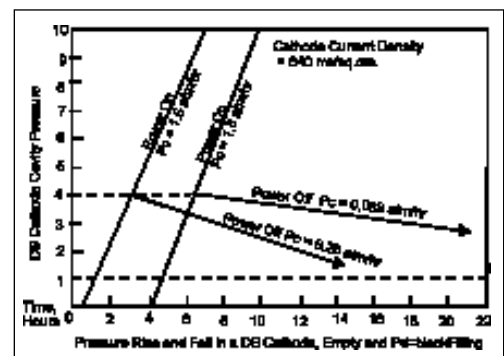
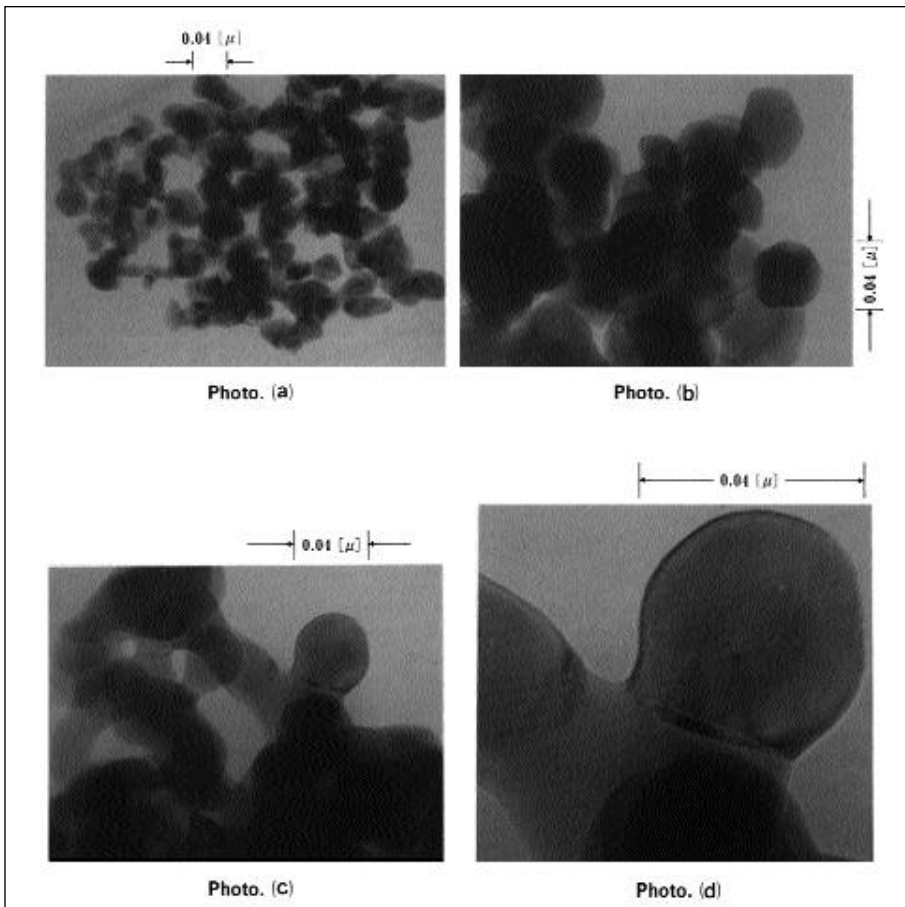


Fig.16 Pressure Rise and Fall in an Empty and Filled DS Cathode



Figures 17 & 18. Comparison between non-deuterated and highly deuterated Pd-black by electron micrograph.
Photo (a) and (b) are non-deuterated sample.
Photo (c) and (d) show the highly deuterated sample.

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3. "New Energy from a Double Structured Cathode," Yoshiaki Arata and Yue-Chang Zhang, 1994. Translation from Japanese by A. Kawasaki.

Figures:

- Fig. 1: Fig. 5, Ref 1
- Fig. 2: Fig. 6b, Ref 1
- Fig. 3: Fig. 8a, Ref 1
- Fig. 4: Fig 19(a), Ref 1
- Fig. 5: Fig 22[III] (A), Ref 1
- Fig. 6: Fig 22[III](A), Ref 1
- Fig. 7: Fig 22[III](Ba), Ref 1
- Fig. 8: Fig 22[III](Bb), Ref 1
- Fig. 9: Fig 2a, Ref 3
- Fig 10: Fig 4(A), Ref 2
- Fig 11: Fig 1(A1), Ref 2
- Fig 12: Fig 1(A3), Ref 2
- Fig 13: Fig 6(B), Ref 2
- Fig 14: Fig C(a), Ref 2
- Fig 15: Fig 7(a), Ref 1
- Fig 16: Fig 6(a), Ref 2
- Fig 17: Photo (a) p40, Ref 1
- Fig 18: Photo (d) p40, Ref 1
- Fig 19: Fig 17, Ref 1

Appendix A: Testing for Tritium

As mentioned above, tritium and heli-

um 3 cannot be distinguished by QMS, for the masses are nearly equal. A&Z looked instead for compounds of tritium by comparing mass spectrometry of deuterated and non-deuterated samples. This work was done before they built the closed QMS system discussed above.

Conventional mass spectrometers use an attenuated flowing gas stream containing a known calibrating gas plus the specimen gas. The gas flows past an ionizing structure and into the exhaust pump. Ionized particles are drawn into the electric and magnetic fields which sort them by charge and mass. Where helium is the species of interest, as in the present case, the large volumes of carrier gas used may well be contaminated with ambient helium, which has led to doubt about claims for helium in F&P-class experiments. Under high vacuum conditions, it is also possible for particles (molecules and atoms) to escape from the vacuum pump inlet and migrate into the vacuum chamber; this is called *back pressure*. Such migration is another pathway for helium contamination.

F&P improved on the conventional mass spectrometer by introducing a controlled leak of a known gas into the inlet

port of the pump. This leak provides a stream of molecules which sweep escaping molecules back into the pump. Some of these leaking molecules can also diffuse to the mass spectrometer, but the species is known and can be neglected in analysis of the results. A&Z refer to this as a "limited QMS." Limited QMS gave A&Z the first look at the mass spectrum of deuterium and helium-4 together. It was also used to produce the data of Fig 19 below.

Because of the complexity of the data in this figure, I have made no attempt to trace or clarify it, but offer a guided tour. What is presented are the intensities of QMS target currents for mass ranges from 1 to 20, taken with highly-deuterated (*) and non-deuterated (open circle) samples. The test period is a bit over 15 minutes. The QMS is operated in a low resolution mode, which shows only the gross features of the spectra, not the details shown elsewhere. Note the extreme dynamic range, over seven orders of magnitude.

For each mass value, possible isotopes and compounds are listed. Most of these show an expected trend with time; a burst of signal as the sample is heated and contaminants burn off, followed by a steadily declining signal as the heating and pumping proceeds.

In this experiment A&Z present the evidence against the presence of tritium in their processed Pd-black samples. Tritium is abundantly produced by the Rutherford reaction and compounds of tritium would be present if produced. These would be at mass 5,6,and 7. Note that the traces for the deuterated and non-deuterated cases overlap at the noise level for these masses, and that the tritium compounds have been "Xed" out. The trends for mass 3 and 4 are different from the others and require closer consideration. Many compounds of H, C, and O are possible assignments for the mass traces. A&Z do not provide an extensive discussion of these, so my analysis is as follows:

Surface contamination of the Pd-black may consist of water and an undefined blend of organics. As the sample is heated up to 1300° C, these will evaporate and dissociate and recombine in the effluent vapor into simpler compounds. Deuterium is present in nature in a concentration of about 0.01% that of hydrogen. Deuterium is thus present even in the non-deuterated samples. The essentially level output with time of the non-deuterated trace at mass 3 is assigned to DH and its source may be from the larger mass of ambient D in the surface contamination which winds up as

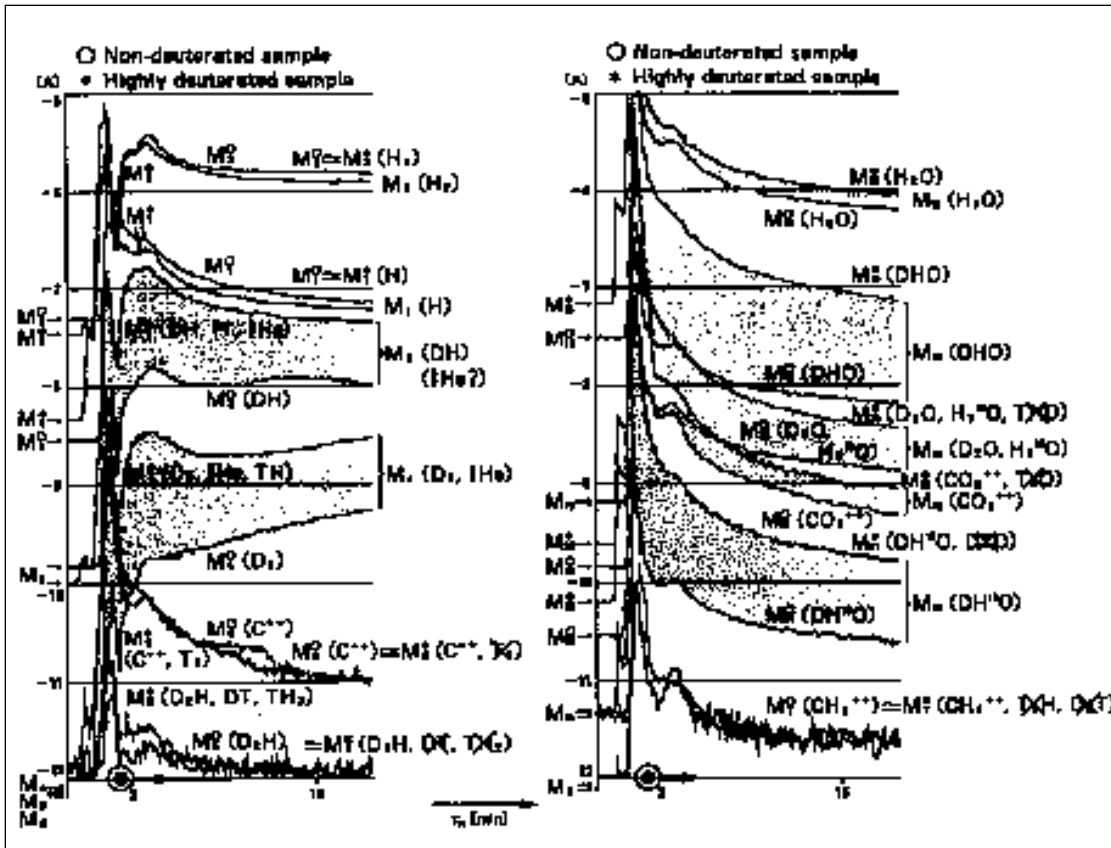


Fig.19. Composite QMS Data for Mass 1-22 for Heavily Deuterated and Non-Deuterated Pd-black Samples as a Function of Time, Using "Limited QMS" Procedure

DH in the chemical reactions involved with the heating. The non-deuterated mass 4 trace rises with time. It is assigned to D_2 and is the expected four orders below the H_2 levels. It rises with time, which may be due to dissociation of larger molecules containing D by the sample heating. Note that for both mass 3 and 4, the highly deuterated samples from the cathode show levels of mass 3 and 4 nearly an order of magnitude higher than for the unprocessed samples.

Appendix B: Determination of $^4\text{He} / ^3\text{He}$ Ratio

Mass 3 contains both ^3He and DH. They can be separated by selection of the ionization potential of the QMS. Below 24.5 V, the hydrogen isotopes and compounds such as DH are ionized. Above 25 V, the helium series, including ^3He and ^4He are ionized. By a series of comparative measurements, A&Z determined that four parts of ^4He are produced for every 3 parts of ^3He . This is also direct evidence against the Rutherford reaction.

