CMNS Research Progressing in Kobe University -Deuterium Permeation and Absorption--

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Experimental studies on condensed matter nuclear science (CMNS) ongoing in Kobe University are reviewed. One is the subject of nuclear transmutation during forced permeation of deuterium (D) through a multi-layered film of X/CaO/Pd, with X being an element to be transmuted, \textit{e.g.}, \textsuperscript{133}Cs transmuted to \textsuperscript{141}Pr, \textsuperscript{88}Sr to \textsuperscript{96}Mo, \textit{etc.}, which was originally claimed by Iwamura \textit{et al.}\textsuperscript{[1]}. In addition to the modified version of the sample exposure system with reversed flow direction installed at a beam line of a tandem electrostatic accelerator 5SDH-2, a stand-alone D permeation system was used to examine the phenomenon by \textit{in-situ} or \textit{ex-situ} PIXE/ERD analysis, respectively.

Using Sr as the element X, more than ten samples having a variety of multi-layer structure have been examined. The PIXE spectra have shown that three out of fourteen samples had increased areal densities of Mo in exchange for decreased those of Sr after D permeation with integrated flow rates up to 2 - 20\times10^{22} \text{ cm}^{-2}. However, the K\alpha-K\beta X-ray peaks in the spectra were too small to give sufficiently high reliability to the identification of the peaks, and to deduce definite conclusion of transmutation. It should be noted, nonetheless, that almost all samples with exception of only one had decreased areal densities of Sr after D permeation.

Tungsten was also tested as the element X. Assuming the regularity in transmutation that (atomic number, mass number) increase by (4, 8), we expected production of radioactive nuclei \textsuperscript{191}Pt (\textit{T}_{1/2} = 2.86 \text{ d}) from \textsuperscript{183}W. An NaI(Tl) scintillation probe and a HPGe detector were employed to detect 0.538-MeV gamma rays from \textsuperscript{191}Pt. The result of the measurement for the first sample has shown that the areal...
The abstract begins with an introduction to the topic, stating that the density of $^{191}$Pt is smaller than $10^7 \text{ cm}^{-2}$, if any. The second subject is to confirm heat and $^4$He generation by D absorption in nano-sized Pd powders reported by Arata and Zhang [2], and to investigate the underlying physics. The research involves installing a twin system to perform calorimetry during $D_2$ or $H_2$ absorption by micronized powders of Si, Pd, Pd-black, and Pd-Zr oxide compounds. The project is performed as a joint research program with Technova Inc., and is described in detail in the following two presentations [3, 4].


Deuterium Gas Charging Experiments with Pd Powders for Excess Heat Evolution

(I) Results of absorption experiments using Pd powders


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Technova Inc.

To confirm heat and $^4$He generation by deuterium (D) absorption in nano-sized Pd powders reported by Arata and Zhang [1], and to investigate the underlying physics, we have installed a twin system of double structured vessels to perform flow calorimetry during $D_2$ or $H_2$ absorption by a variety of micronized Pd samples. The inner reaction vessels contain sample powders of Si, Pd, Pd-black, or Pd-Zr oxide compounds, and have calorimetry arrangement, i.e., coolant pipes and thermocouples on the outer surfaces. The inner vessels are located in the outer chambers which are evacuated for thermal insulation. We can perform $D_2$ and $H_2$ absorption runs for two samples at the same time under the same environmental condition. Nuclear diagnostic tools, a 2”×2” NaI(Tl) scintillation probe and a neutron dose-rate meter, are also provided around the system.

The first-stage experiments are described in detail in ref. [2]. The evolution of pressure and temperature after introduction of 1-MPa $D_2/H_2$ gas was divided into two phases. In the first phase, during which the pressure was kept well below 10 kPa, the D (H) loading ratio of D/Pd (H/Pd) and the specific output energy reached 0.85 (0.78) and $0.54 \pm 0.10$ (0.45±0.08) kJ/g-Pd, respectively, for the Pd-black, which were about 2 – 3 times greater than those for the 0.1-$\mu$m-Pd powder. Moreover, when
D$_2$ gas was used with Pd-black, apparent excess heat production in the second phase was implied, although temperature oscillations and drift were too large to confirm the result.

Then in the second stage, the system was modified to improve the accuracy: The heat capacity of the reaction vessel was decreased, while increasing the mass of the test sample, to minimize the time constant of the calorimeter and maximize the sensitivity.

First, repeated use of the Pd-black samples after one or more cycles of D (H) absorption-degassing has revealed the Pd-black powder stuck with each other to become larger micro-particles without nanoscale structure. Next, nano-sized powders of mixed Pd and Zr oxides fabricated by Santoku Cooperation, Kobe, Japan, have been used to reveal their interesting and exciting characteristics, which is discussed in detail in the succeeding presentation [3].


Deuterium Gas Charging Experiments with Pd Powders for Excess Heat Evolution
(II) Discussions on Experimental Results and Underlying Physics

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$^2$(Division of Marine Engineering, Graduate School of Maritime Sciences, Kobe University)

Our experimental results [Ref-1] obtained for Pd/PdO/ZrO$_2$ nano-composite samples under the joint research of Kobe University and Technova Inc. in the 2008 fiscal year are summarized as:

1) Arata-Zhang’s excess heat result demonstrated in May 2008 was replicated quantitatively.
2) For Pd/PdO/ZrO$_2$ powders (produced by Santoku Co., Kobe Japan):
   2-1) D-gas charge in the 1$^{\text{st}}$ phase (zero pressure interval) gave 20–90 % excess heat than H-gas charge.
   2-2) In the 2$^{\text{nd}}$ phase of pressure rise (finally up to 1MPa), significant excess heat (about 2 kJ/g-Pd) for D-gas charge was observed, while near zero level excess heat for H-gas charge was observed.
3) No increase of neutron counts was seen, nor increase of gamma-ray counts.
4) D/Pd loading ratio in the end of 1$^{\text{st}}$ phase was 1.0, while H/Pd ratio was about 0.8-0.9.
5) Released energy per D or H-atom in the 1$^{\text{st}}$ phase was 1.7-1.8eV or 1.0-1.6eV respectively for D or H. These specific values are anomalously large, compared with known values of 0.5eV and 0.2eV per D or H respectively for surface-adsorption and lattice-absorption of hydrogen gas into bulk Pd
JCF9 abstract

metal.

6) After evacuation, used Santoku sample retains 100 times more deuterium than Pd-black sample, seen by degassing data by baking up to 574 K.

7) In repeated run with used sample after #1, we can expect further steady excess heat. We observed similar effect as “heat after death” after evacuation of reaction chamber in the end of run.

For the 1st phase, we discuss the underlying surface and nano-particle physics in views of the enhanced surface adsorption potential by fractal sub-nano-scale trapping points on nano-Pd particle, the diffusion to inner shallower Bloch potential of regular Pd lattice, and the drastic mesoscopic and isotopic effect of surface and lattice rearrangement of nano-Pd particle by full D(H)-absorption to make deeper D(H) trapping potentials of surface adsorption (about 2eV for D) and intermediate surface state trapping.

We add discussions on the 2nd phase where we observed long lasting significant excess heat only for D-charging, in the view of possible 4D/TSC model type “condensed cluster fusion reaction” to produce excess heat and 4He particles by preferred 4D fusion without neutrons and gamma rays.


[ACS 2009 NET Session Abstract]

Deuterium gas charging experiments with Pd powders for excess heat evolution

ENVR 51

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We have started a series of deuterium (and hydrogen) gas charging experiments with Pd nano-powders to study possible heat evolution and D (or H)-loading characteristics by using a revised Arata-type twin system. The twin system is made of identically designed A1 and A2 systems, in each of which an inner gas-charging cell with flow calorimeter and an outer vacuum chamber are set up. The A1 system is used for D-gas foreground run, and the A2 system is for the H-gas blank run. Our first data with two commercially available Pd powders (0.1 micron Pd particles and Pd-black) are already meaningful. Experiments with Pd-black sample gave 2.6 kJ/g-Pd excess heat for the second phase of 1,300 minutes operation and D/Pd=0.85 for the first phase (about 100 min interval from start) with zero D-gas pressure. No excess heat with H-gas charging was seen with H/Pd=0.78. Experiments with 0.1 micron Pd powders gave D/Pd =0.45 for the first phase and much less excess heat for the second phase. We are extending experiments for nano-fabricated Pd samples to be reported at the meeting. In situ radiation monitors are for neutron and gamma-ray. Elemental analysis of "before/after" samples is done by PIXE. 4He detection will be also tried.
Deuterium Gas Charging Experiments with Pd Powders for Excess Heat Evolution

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A. Takahashi, R. Seto, and Y. Fujita
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to be presented at ACS2009 NET Session
Salt Lake City
March 22-26, 2009
It has been recently reported in ref. [1] that charging of **highly pure D$_2$ gas** into Pd nano-powders in the form of Pd/ZrO$_2$ nano-composite contained in a stainless-steel vacuum vessel has induced significant excess heat and $^4$He generation.

We have constructed an experimental system to replicate the phenomenon of **excess-heat** (and $^4$He generation) and **investigate the underlying physics for D(H)-charged Pd powders**.

We constructed two identical chambers (twin system); one for $\text{D}_2$ gas foreground run and the other for $\text{H}_2$ gas background run. Each system has an inner reaction chamber containing Pd powders (Pd-black and nano-Pd/ZrO$_2$) and the outer chambers are evacuated for thermal insulation.

A water-cooling system is provided for flow calorimetry to estimate heat production rates for two phases.

D(H)/Pd ratios were measured for the 1$^{\text{st}}$ phase (“zero pressure interval”).

Moreover, after the gas charging, elemental analysis of the Pd powder was performed by PIXE (Particle Induced X-ray Emission) analysis. Radioactivity was measured with a Hp-Ge detector. A REM counter was used for monitoring neutron emission. NaI scintillator was used for gamma-ray. Furthermore, $^4$He analysis will be performed in the future.
Reduced view of the system

- D₂ gas cylinder
- H₂ gas cylinder
- Vacuum pumps
- Pressure gauge
- Vacuum gauge
- Reaction chamber
- Outer vacuum chamber

A₁ system

A₂ system
1. Calorimetry Calibration

Outline

The temperature of the reaction chamber is thermally isolated by evacuating the outer vacuum chamber.

Flow calorimetry is provided to measure heat output from the sample cup in the reaction chamber.

Calibration of the calorimetry system

• Input power: 0.5W, 1W, 3W, 6W and 10W.
• D₂ gas pressure in the reaction chamber: 0, 0.1, 0.3, and 1.0 MPa.
• Flow velocity of coolant water: 6 ml/min.
Heat recovery rate is independent of pressure and input power, and is about **70% (±2%)**.
Thermal time constant: temperature change following the change in the input power.

\[
\Delta T = \exp\left(-\frac{t}{\tau_1}\right) + \exp\left(-\frac{t}{\tau_2}\right)
\]

<table>
<thead>
<tr>
<th>MPa</th>
<th>(\tau_1) [s]</th>
<th>(\tau_2) [s]</th>
</tr>
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<tbody>
<tr>
<td>0</td>
<td>1500</td>
<td>2700</td>
</tr>
<tr>
<td>0.1</td>
<td>690</td>
<td>3000</td>
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<tr>
<td>0.3</td>
<td>750</td>
<td>2600</td>
</tr>
<tr>
<td>1.1</td>
<td>930</td>
<td>2000</td>
</tr>
</tbody>
</table>

Thermal time constants will be used to determine the phases in absorption runs.
The Pd membrane (0.2 mm-t, 99.95%) separates the evacuated reaction chamber (1.6 ℓ) and the gas reservoir filled with $D_2$ at 1 MPa.

Permeation rate of $D_2$ gas into the reaction chamber was derived as a function of membrane temperature by measuring the rate of pressure increase after stopping evacuation.

$D_2$ gas flow rate is controllable between 0.1 and 25 sccm by varying the temperature from 298 K to 900 K.
Pd powders ($\phi0.1\mu$m, 99.5%) – results

The evolution is divided into 2 phases by the time 30 minutes after the beginning of the pressure rise.

The release of heat of hydride formation should be completed in the 1st phase.
Likely to the case of Pd φ0.1μm sample, the evolution looks similar. The evolution can be divided into 2 phases by the time 30 minutes after the beginning of the pressure rise. The release of heat of hydride formation should be completed in the 1st phase.
B. Pd black (300 mesh, 99.9%) – comparison of Heat balance

- The output energies in the 1st phase are almost the same for both cases, but seem to be somewhat larger than the nominal values of 100 – 405 J/g (H₂) and 80 - 330 J/g (D₂).
- On the contrary, the output energy in the 2nd phase appears to be larger for D₂ than H₂.
(1) D (H) absorption rates -D/Pd or H/Pd- for Pd black were about twice of those for Pd φ0.1μm for the phase 1 charging.

(2) Apparent excess heat about 2.6 kJ/g was observed for Pd black with D₂ gas charging.
Summary Remarks for the 1st Series Experiments

- The Pd-H$_2$/D$_2$ gas absorption system using commercial Pd powder has been installed which enables a flow calorimetry and nuclear diagnosis.

- The calibration examination and the first experimental results have been described.

- When D$_2$ gas was used for the Pd-black sample, trend of excess heat was observed.

- Calorimetry time constant is yet to be made small by making heat capacity of the reaction chamber small.

- Clearer results of heat evolution will be obtained also by increasing the amount of samples.

- Nano-sized Pd powders will also be examined.
The 2\textsuperscript{nd} Series Experiments

New A: Shorter FC Time-Constant (5min) and Larger Cell Volume
Fig. 4-2. Evolution of temperature; D₂ gas contained with 1MPa in the reaction chamber.
Time Constant for Heat Removal

近似式より熱時定数

- 0MPa; \( \tau = 5.58 \text{ (min)} \)
- 0.3MPa; \( \tau = 5.28 \text{ (min)} \)
- 1.0MPa; \( \tau = 4.85 \text{ (min)} \)

Old A-System

<table>
<thead>
<tr>
<th>MPa</th>
<th>Thermal time constant ( \tau_1 ) [s]</th>
<th>Thermal time constant ( \tau_2 ) [s]</th>
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<tbody>
<tr>
<td>0.0</td>
<td>1463</td>
<td>2731</td>
</tr>
<tr>
<td>0.1</td>
<td>686</td>
<td>3016</td>
</tr>
<tr>
<td>0.3</td>
<td>746</td>
<td>2566</td>
</tr>
<tr>
<td>1.1</td>
<td>927</td>
<td>2048</td>
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</table>

New A-System

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<th>MPa</th>
<th>Thermal time constant ( \tau ) [s]</th>
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</thead>
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<td>0.0</td>
<td>335</td>
</tr>
<tr>
<td>0.3</td>
<td>317</td>
</tr>
<tr>
<td>1.0</td>
<td>291</td>
</tr>
</tbody>
</table>
2. Experimental Procedure

Set Sample in Inner Cell

Evacuation and Baking (<300°C)

Start D (H) Gas Charge Run #1

Take Data: Calorimetry, Pressure, Neutron, Gamma-ray, etc.

Evacuation and Baking (<300°C) Take De-gas Data

Start D (H) Gas Charge Run #2

Take Data: Calorimetry, Pressure, Neutron, Gamma-ray, etc.
Pd-black (20g) / D-gas charge #1: Nov. 2009

- Heat output
- Pressure

D/Pd = 0.78

New gas cylinder

Power (W)
Pressure (MPa)

Time (min)

1st phase
2nd phase
Pd-black / D-gas charge #1 data

Power (W)

Heat -peak

Pressure

Rearrangement Here?
Decreased absorption and heat level for #2

Pd-black (20g)/ D-gas #2

Heat output

Pressure

D/Pd=0.23

Heat/ Power

Time

1st phase

2nd phase

時間(min)

出力(W), ΔT(K)

(W)

(MPa)
After experiments, Pd black powders stuck to be bigger sizes → Decrease of active surface area
Before experiment, surface was fractal in nano-scale.

After experiment, surface became flatter in about 10 times larger scale → Decrease of active surface area
Summary for Pd-black

- There were seen excess heat in the 1\textsuperscript{st} and 2\textsuperscript{nd} phases, but smaller than those by Pd/ZrO\textsubscript{2}.
- Pd-black powders stuck to be bigger sizes (about 10 times diameter by SEM observation), after the #1 run.
- In the #2 run and following runs, no significant excess heat was seen.
- D/Pd ratio for #1 was 0.78 in the pressure zero condition under D-charge.
- D/Pd ratios for #2 and later runs, D/Pd was about 0.23 (much smaller).
- No transmuted elements were seen by PIXE.
Constant Flow Pump
Const. Temp. Water Bath
Chiller Unit
Cold Trap
D₂ gas
H₂ gas

Constant Flow Pump
Const. Temp. Water Bath
Chiller Unit
Cold Trap
D₂ gas
H₂ gas
A1 System
Experiments with Pd/PdO/ZrO$_2$ Dispersed Samples

10g (Net Pd weight : 4.3 g): Three trials for Santoku 1, Santoku 2 and Santoku 3 samples, #1 and #2 runs for each sample

• Nano-Pd/ZrO$_2$ sample was produced by Santoku Co. Japan, based on different protocol from Inoue-Yamaura (Arata-Zhang).

• X-ray diffraction analysis showed composite of Pd/PdO/ZrO$_2$.

• Pd particle size is less than 10nm.

• A1 system: 10g for D-gas charging (2-3sccm)

• A2 system: 10g for H-gas charging (2-4sccm)

• Water-flow calorimetry: 6cc/min flow rate
Santoku 1#1 Results of Heat Evolution

Gas flow rate: 2.8 (D) and 2.6(H) sccm

Output power for D2 run
Pressure for D2 run
Output power for H2 run
Pressure for H2 run

Power by D-gas (A1)
Power by H-gas (A2)
Santoku 1#1 Expanded View
For 1st phase

A2 first phase end:
H/Pd=0.9

A1 first phase end:
D/Pd=1.0

Heat (D): A1

Heat (H): A2 x0.5

7.0 kJ→1.6kJ/g-Pd→1.8eV/D

3.6 kJ→1.3kJ/g-Pd→1.0eV/H

Power

0.0

0.2

0.4

0.6

0.8

1.0

出力(W)

Time 時間(min)
Results of Santoku2#1 run

A1: D-gas Charge with 2.9sccm

A2: H-gas Charge with 3.5sccm
Comparison of Results for Santoku1#1 and Santoku2#1
Pd · ZrO$_2$ Santoku 2 #1 Run (A1) for D-gas Charge

Output power [W]
Output power for D$_2$
Pressure for D$_2$

1.5kJ/g-Pd → 1.7eV/D

1.6kJ/g-Pd

D/Pd=0.97

1st phase

2nd phase
**Pd · ZrO₂ Santoku 2 #1 Run (A2) for H-gas Charge**

![Graph](Image)

- **Output power for H₂ run**
- **Pressure for H₂ run**

**Key Points**:
- **1.2 kJ/g-Pd → 1.6eV/H**
- **H/Pd=0.81**
- **0.4kJ/g-Pd**

**Graph Details**:
- The graph shows the relationship between time and output power for the H₂ run.
- The pressure for the H₂ run is also depicted.
- The graph is divided into two phases:
  - **1st phase**: Initial rapid increase in output power.
  - **2nd phase**: Steady state output power.

**Additional Information**:
- Noise is indicated in the graph.

**ACS09**
**Integrated Data for Pd/ZrO$_2$ Santoku #1 Runs**

Significant **isotopic effects** are seen for D(H)/Pd = $x$ loading ratios, and output energies per Pd (and D(H) atom), for the 1$^{st}$ phase.

Significant **excess heat** by D-gas charge in the 2$^{nd}$ phase.

### Santoku 2#1

<table>
<thead>
<tr>
<th>Run number</th>
<th>Specific output energy [kJ/g]</th>
<th>Composition; $x$ for</th>
<th>Output energy per D/H</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>1st phase</td>
<td>2nd phase</td>
<td></td>
</tr>
<tr>
<td>D$_2$</td>
<td>1.5±0.0</td>
<td>1.6±0.2</td>
<td>0.97</td>
</tr>
<tr>
<td>H$_2$</td>
<td>1.2±0.0</td>
<td>0.4±0.2</td>
<td>0.81</td>
</tr>
</tbody>
</table>

### Santoku 1#1

<table>
<thead>
<tr>
<th></th>
<th>Specific output energy [kJ/g]</th>
<th>Composition; $x$ for</th>
<th>Output energy per D/H</th>
</tr>
</thead>
<tbody>
<tr>
<td>D$_2$</td>
<td>1.6±0.0</td>
<td>1.7±0.2</td>
<td>1.0</td>
</tr>
<tr>
<td>H$_2$</td>
<td>0.8±0.0</td>
<td>(-)</td>
<td>0.89</td>
</tr>
</tbody>
</table>
Summary Table of Integrated Data for #1 Runs With Santoku Pd/PdO/Zr₂O nano-composite samples

<table>
<thead>
<tr>
<th>Run number</th>
<th>flow rate [sccm]</th>
<th>Output energy [kJ]</th>
<th>Specific output energy [kJ/g]</th>
<th>Composition: x for PdDₓ/PdHₓ</th>
<th>Output energy per D/H in the 1st phase [eV]</th>
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<tbody>
<tr>
<td>D-PZ1#1</td>
<td>2.8</td>
<td>7.0±0.2</td>
<td>6.8±1.3</td>
<td>1.6±0.0</td>
<td>1.6±0.3</td>
</tr>
<tr>
<td>Corrected H-PZ1#1</td>
<td>2.6</td>
<td>3.6±0.1</td>
<td>-5.1±1.4</td>
<td>0.8±0.0</td>
<td>-1.2±0.3</td>
</tr>
<tr>
<td>D-PZ2#1</td>
<td>2.9</td>
<td>6.4±0.2</td>
<td>5.5±0.8</td>
<td>1.5±0.0</td>
<td>1.2±0.2</td>
</tr>
<tr>
<td>H-PZ2#1</td>
<td>3.5</td>
<td>5.1±0.1</td>
<td>1.7±0.9</td>
<td>1.2±0.0</td>
<td>0.4±0.2</td>
</tr>
</tbody>
</table>

Time-Interval: 0-1,800 min
Arata-Zhang’s 1st Phase Data for Pd/ZrO₂ (7g)

Arata-Zhang:  \[\text{Heat}(D)/\text{Heat}(H) \sim (72\text{C}-25\text{C})/(60\text{C}-25\text{C}) = 1.34\]

Our Exp. Santoku1:  \[\text{Heat}(D)/\text{Heat}(H) = (7.0kJ)/(3.6kJ) = 1.94\]

Our Exp. Santoku2:  \[\text{Heat}(D)/\text{Heat}(H) = (6.4kJ)/(5.1kJ) = 1.26\]

Agreed!
Results by Large H-Flow Rate And Low D-Gas Pressure

Santoku 3#1 Run

A1: H-gas, Flow Rate = 9.96 sccm
A2: D-gas, Flow Rate = 2.1 sccm

<table>
<thead>
<tr>
<th>run</th>
<th>1st phase [kJ]</th>
<th>2nd phase [kJ]</th>
<th>1st phase [J/g]</th>
<th>2nd phase [J/g]</th>
</tr>
</thead>
<tbody>
<tr>
<td>D₂-3-1</td>
<td>7.14±0.15</td>
<td>1.26±1.36</td>
<td>1661±35</td>
<td>293±316</td>
</tr>
<tr>
<td>H₂-3-1</td>
<td>7.07±0.07</td>
<td>-0.23±1.44</td>
<td>1644±18</td>
<td>-54±334</td>
</tr>
</tbody>
</table>
# Table: Integrated Data for the First Phase ("zero" pressure interval)

<table>
<thead>
<tr>
<th>Run</th>
<th>gas</th>
<th>Heat (kJ/g-Pd)</th>
<th>D/Pd or H/Pd</th>
<th>Eout/Pd (eV)</th>
<th>Eout/D(H) (eV)</th>
<th>comment</th>
</tr>
</thead>
<tbody>
<tr>
<td>0.1 μ Pd#2</td>
<td>D</td>
<td>0.097±0.05</td>
<td>0.43</td>
<td>0.11</td>
<td>0.25±0.13</td>
<td>Old A</td>
</tr>
<tr>
<td>ibid</td>
<td>H</td>
<td>0.12 ±0.03</td>
<td>0.45</td>
<td>0.13</td>
<td>0.29±0.07</td>
<td>Old A</td>
</tr>
<tr>
<td>Pd-black(I)</td>
<td>D</td>
<td>0.54 ±0.10</td>
<td>0.85</td>
<td>0.59</td>
<td>0.70±0.13</td>
<td>Old A</td>
</tr>
<tr>
<td>ibid</td>
<td>H</td>
<td>0.45 ±0.08</td>
<td>0.78</td>
<td>0.50</td>
<td>0.63±0.11</td>
<td>Old A</td>
</tr>
<tr>
<td>Pd-black(II)#1</td>
<td>D</td>
<td>0.47 ±0.06</td>
<td>0.78</td>
<td>0.52</td>
<td>0.66±0.08</td>
<td>Baking T(\leq 170°C)</td>
</tr>
<tr>
<td>Pd-black(II)#2</td>
<td>D</td>
<td>0.17 ±0.03</td>
<td>0.23</td>
<td>0.19</td>
<td>0.81±0.14</td>
<td>stuck</td>
</tr>
<tr>
<td>Pd-black(II)#4</td>
<td>D</td>
<td>0.16 ±0.02</td>
<td>0.24</td>
<td>0.18</td>
<td>0.75±0.09</td>
<td>stuck</td>
</tr>
<tr>
<td>Pd-black(II)#2</td>
<td>H</td>
<td>0.16 ±0.01</td>
<td>0.22</td>
<td>0.18</td>
<td>0.80±0.05</td>
<td>stuck</td>
</tr>
<tr>
<td>Santoku 1 #1</td>
<td>D</td>
<td>1.63 ±0.01</td>
<td>1.0</td>
<td>1.80</td>
<td><strong>1.80±0.01</strong></td>
<td>Baking T(\leq 3000°C)</td>
</tr>
<tr>
<td>Pd/PdO/ZrO(_2)</td>
<td>D</td>
<td>0.80 ±0.01</td>
<td>0.89</td>
<td>0.91</td>
<td><strong>1.02±0.02</strong></td>
<td>ibid</td>
</tr>
<tr>
<td>ibid</td>
<td>H</td>
<td>1.49 ±0.01</td>
<td>0.97</td>
<td>1.70</td>
<td><strong>1.71±0.02</strong></td>
<td>ibid</td>
</tr>
<tr>
<td>Santoku 2 #1</td>
<td>D</td>
<td>1.30 ±0.01</td>
<td>0.81</td>
<td>1.48</td>
<td><strong>1.67±0.02</strong></td>
<td>ibid</td>
</tr>
<tr>
<td>ibid</td>
<td>H</td>
<td>1.66 ±0.03</td>
<td>0.77</td>
<td>1.89</td>
<td><strong>2.38±0.02</strong></td>
<td>A2, 2.1sccm</td>
</tr>
<tr>
<td>Santoku 3 #1</td>
<td>D</td>
<td>1.64 ±0.02</td>
<td>1.57</td>
<td>1.87</td>
<td><strong>1.15±0.02</strong></td>
<td>A1, 10sccm</td>
</tr>
</tbody>
</table>

Note-1: \(\text{H}_2\text{O} \rightarrow 2\text{H} + \text{O} \rightarrow 1.48\text{eV}\), and \(\text{D}_2\text{O} \rightarrow 2\text{D} + \text{O} \rightarrow 1.52\text{eV}\) \((0.75 \text{ eV per H(D)})\)

Note-2: \(\text{Eout}/\text{D(H)}\) values for chemical heat are 0.6 to 0.8 eV per D (or H), for nano-powders of Pd-black

Note-3: \(\text{Eout}/\text{D}\) value 1.7-2.3eV for Santoku-nano-Pd powder anomalously large. And \(\text{Eout}/\text{H}\) values are also anomalously large compared to known chemical values.

Note-4: Pd-black powder stuck to be bigger sizes after #1 run, but heat/D(H) is same to show some surface reaction active.
Discussions for the 1\textsuperscript{st} Phase

- Pd-black: 
  \[(\text{Heat/D})_{\text{av}} = 0.70 \pm 0.15 \text{ eV} \]
  \[(\text{Heat/H})_{\text{av}} = 0.69 \pm 0.1 \text{ eV} \]

- Santoku1: 
  \[(\text{Heat/D}) = 1.80 \pm 0.02 \text{ eV (2.38 for Santoku3\#1)} \]
  \[(\text{Heat/H}) = 1.02 \pm 0.02 \text{ eV (1.15 for Santoku3\#1)} \]

- Santoku2: 
  \[(\text{Heat/D}) = 1.71 \pm 0.02 \text{ eV} \]
  \[(\text{Heat/H}) = 1.67 \pm 0.02 \text{ eV (* flow rate larger for H-gas)} \]

- After Fukai book: \(0.2\text{eV/H for bulk H absorption.}\)
  \(100\text{kJ/mol-H}_2 : 0.5\text{eV/H for surface adsorption.}\)

- Reaction may be Surface Mesoscopic Phenomenon for the 1\textsuperscript{st} Phase (“zero pressure” interval). Isotopic effect is visible.

- Pd nano-particle makes deep trapping potential of D(H), probably in fractal defects of its surface!?

- This is the reason of high loading “in vacuum” for Pd-nano.
After evacuation, Santoku sample retains much more (100 times) D(H) than Pd-black: due to mesoscopic effect (rearrangement of surface and lattice) of Pd nano-particles.

4-3. Variation of pressure during sample baking for outgassing.
Metal-Oxide-Nano-Pd Composite

- PdDx
- $X=1.0$ by Arachi et al.; D-Absorption in O-sites of Pd-Lattice (We support this data)
- $X=2.0$ by Yamaura et al.; deuterons at Pd-ZrO$_2$ interface (voids?)
- Arata claimed: $x=2.5$

Pd Nano-Particle
About 5 nm mean diameter

About 10 micron

ZrO$_2$
Irregular and Fractal Sites should form on surface of nano-particle

5 nm Pd particle: 4,000-6,000 atoms

D$_2$ molecule

Fractal Trapping points
Potential form of hydrogen adsorption and absorption near surface

For nano-holes:
The heavier is the isotope, the more enhanced adsorption takes place on surface.

Periodical Lattice Potential
0.23eV deep for Pd
Esol = 0.032 eV
Long time lasting heat by D-charge without input power:
29.2kJ Total by 24.4g Pd/ZrO$_2$ sample
### Table 1: Run number, Composition; x for PdDx/PdHx, Output energy per D/H in the 1st phase [eV]

<table>
<thead>
<tr>
<th>Run number</th>
<th>flow rate [sccm]</th>
<th>Output energy [kJ]</th>
<th>Specific output energy [kJ/g]</th>
<th>Composition; x for PdDx/PdHx</th>
<th>Output energy per D/H in the 1st phase [eV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>D-PZ1#1</td>
<td>2.8</td>
<td>7.0±0.2</td>
<td>1.6±0.0</td>
<td>1.0</td>
<td>1.80 ± 0.05</td>
</tr>
<tr>
<td>Corrected H-PZ1#1</td>
<td>2.6</td>
<td>3.6±0.1</td>
<td>-5.1±1.4</td>
<td>0.89</td>
<td>1.02 ± 0.03</td>
</tr>
</tbody>
</table>
Discussions on Total Excess Heat Rates for #1 Runs: Our results are comparable to Arata’s.

- **Arata-Zhang Exp.**
  - Sample Pd/ZrO$_2$ by Fukuda P. M. Co.
    - weight: 24.4 g
    - net Pd weight: 10.5 g
  - Observed Excess Heat (0-3000min): 29.2 kJ
  - Excess Heat Rate: $2.77 \text{ (kJ/g-Pd)}$
    (Anomalous heat by H about 1 kJ/g inclusive)
  → Net Value $\sim 1.8 \text{ (kJ/g-Pd)}$

- **Our Exp.**
  - Sample Pd/PdO/ZrO$_2$ by Santoku Co.
    - weight: 10 g
    - net Pd weight: 4.3 g
  - Observed Excess Heat (Example for S2, 0-3000min)
    $S2: (6.4-5.1)+7.0 = 8.3 \pm 1.2 \text{ kJ}$
  - Excess Heat Rate:
    $S1: 2.64 \pm 0.30 \text{ (kJ/g-Pd)}$
    $S2: 1.93 \pm 0.30$
Long Time Lasting Excess Heat by Run: Santoku 2#2

<table>
<thead>
<tr>
<th>run</th>
<th>Gas</th>
<th>Measured flow rate [sccm]</th>
<th>1st phase [kJ]</th>
<th>2nd phase [kJ]</th>
<th>1st phase [J/g]</th>
<th>2nd phase [J/g]</th>
<th>D/Pd or H/Pd</th>
</tr>
</thead>
<tbody>
<tr>
<td>D₂-2-2</td>
<td>D</td>
<td>3.87</td>
<td>0.17±0.03</td>
<td>9.89±1.48</td>
<td>40±7.0</td>
<td>2300±345</td>
<td>0.47</td>
</tr>
<tr>
<td>H₂-2-2</td>
<td>H</td>
<td>3.62</td>
<td>0.58±0.05</td>
<td>1.68±1.46</td>
<td>136±10.9</td>
<td>391±341</td>
<td>0.28</td>
</tr>
</tbody>
</table>
Run: Santoku 2#2: Expanded View of Early –Time Evolution
Run: Santoku 2#2: Expanded View of Heat Evolution after Evacuation- “Heat after Death”

Is Evacuation STIMULUS?
Model b) may correspond to I.

Model a) may correspond to II.

4D/TSC → \(^4\text{He} + \(^4\text{He} + 47.6\text{MeV}\)}
Result of Dynamic Condensation of 4D/TSC by Langevin Equation

1) TSC forms

2) Minimum TSC

3) $^8\text{Be}^*$ formation

4) Break up to two $^4\text{He}$

Electron

Deuteron

Electron Center

$1.4007 \text{ fs}$

$4r_e = 4 \times 2.8 \text{ fm}$
No increase of neutron count rate by REM counter was seen, gamma-ray either.
Conclusions-1

• Arata-Zhang’s Excess Heat Result was replicated quantitatively.

• For Pd/PdO/ZrO₂ powder (Santoku):
  1) D-gas charge in the 1ˢᵗ phase (zero pressure) gave 20-90% excess heat than H-gas charge.
  2) In the 2ⁿᵈ phase, significant excess heat (about 2 kJ/g-Pd) for D-gas charge, while zero level for H-gas charge.

• No increase of neutron counts was seen.

• D/Pd ratio in the end of 1ˢᵗ phase was 1.0, while H/Pd was about 0.8 - 0.9. Flow rate dependence.

• Further experiments changing conditions will be fruitful for developing clean energy devices.
Conclusions

• Nano-Pd dispersed sample (Santoku, Pd/ZrO$_2$) retained 100 times more D(H) atoms after evacuation, than the Pd-black case.

• **Mesoscopic effect by Pd-nano-particle**, namely surface and lattice rearrangement probably makes **deep D(H) trapping potentials (1.0-2.3eV)**. **We need study for D(H)-gas flow-rate dependence.**

• Stable excess heat production is expected for #2 and later runs. **We need further study for this.**

• Detection of nuclear products by the B-system is expected.

• Replication by other groups is important.