Anomalous Heat Generation in Charging of Pd Powders with High Density Hydrogen Isotopes (I) Results of absorption experiments using Pd powders


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To confirm heat and 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 D₂ or H₂ absorption by a variety of micronized Pd samples.

The first-stage experiments are described in detail in ref. [2]. The evolution of pressure and temperature after introduction of 1-MPa D₂/H₂ gas was divided into two phases. The first phase is zero-pressure interval, and in the second phase, pressure increases up to the stationary value. When D₂ 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.

Nano-sized powders of mixed Pd and Zr oxides fabricated by Santoku Corporation, Kobe, Japan, have been used to reveal their interesting and exciting characteristics. In the 1st phase, D-gas charge gave 20~90% excess heat compared to H-gas charge. In the 2nd phase, significant excess heat (about 2 kJ/g-Pd) for D-gas charge was observed, in contrast to near zero level output for H-gas charge. We will further examine the dependence of the anomalous excess heat on the experimental conditions such as the gas flow rate and the sample temperature. The anomalies and the possible mechanisms will be discussed in more detail in the succeeding presentation [3].


Anomalous Heat Generation in Charging of Pd Powders with High Density Hydrogen Isotopes

(I) Results of absorption experiments using Pd powders

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Aim

It has been reported in ref. [1] that charging of highly pure D2 gas into Pd nano-powders in the form of Pd/ZrO2 nano-composite contained in a stainless-steel vacuum vessel has induced significant excess heat.

we have constructed an experimental system to confirm the phenomenon of heat and $^4$He generation by calorimetry and investigate the underlying physics.

Reduced view of the twin system A1A2

- **D$_2$ gas cylinder**
- **A$_2$ system**
- **H$_2$ gas cylinder**
- **A$_1$ system**
- **D$_2$ run**
- **H$_2$ run**
- **Reaction chamber**
- **Outer vacuum chamber**
- **Pressure gauge**
- **Vacuum gauge**
- **Vacuum pumps**
Functional view of the $A_1A_2$ system

- Vacuum pump
- ‘Super’ needle valve
- Cold trap
- D$_2$ or H$_2$

Sample
- Heater

Evacuation
- Baking (340K 3h)
- D$_2$(H$_2$) gas charging

Data acquisition
- Temperature
- Pressure
- Neutron
- Gamma-ray etc.

Experimental procedure

Sample set-up

Evacuation

Baking (340K 3h)

D$_2$(H$_2$) gas charging

Data acquisition
- Temperature
- Pressure
- Neutron
- Gamma-ray etc.

Chiller
Performance of calorimetry

Time resolution: 5 min

Accuracy: ± 14 mW

\[\exp\left(-\frac{t}{\tau}\right)\]
Samples

- $\phi 100$ nm Pd (PP); This is a Pd powder diameter of particle is 100 nm, purity is 99.5%.

- Pd-black (PB); This is a 300 mesh powder and purity is 99.9%

- Santoku Pd (PZ); This is a nano-sized (8 nm and 10.5 nm) powder of mixed-oxides of Pd and Zr (fabricated by Santoku Corporation)
TEM Image of Santoku Pd (10.5 nm)

(By courtesy of the Nuclear Science and Engineering Institute and Particulate Systems Research Center at the University of Missouri-Colombia; Prof. R. Duncan et al.)
The diagram shows the output power and pressure over time for different phases and gases. The following data is presented:

**1st phase**
- D$_2$: 0.10 kJ/g-Pd
- H$_2$: 0.08 kJ/g-Pd
- D/Pd = 0.43
- H/Pd = 0.45

**2nd phase**
- D$_2$: 0.79 kJ/g-Pd
- H$_2$: 0.53 kJ/g-Pd

Flow rates:
- D: 3.5 sccm
- H: 4.3 sccm

The graph illustrates the output power and pressure changes over time for different phases and gases, with a focus on D$_2$ and H$_2$.
Pd-black (PB)

Output power [W] vs. Time [min]

- Blue line: Output (D₂)
- Red line: Output (H₂)
- Purple line: Pressure (D₂)
- Pink line: Pressure (H₂)

1st phase:
- D₂: 0.54 kJ/g-Pd
- H₂: 0.45 kJ/g-Pd
- D/Pd = 0.85
- H/Pd = 0.78

2nd phase:
- D₂: 0.65 kJ/g-Pd
- H₂: -0.62 kJ/g-Pd
- D/Pd = 0.85
- H/Pd = 0.78

Flow rate:
- D: 3.5 sccm
- H: 5.6 sccm
**Santoku Pd ( PZ1,2#1 )**

1st phase

- D2: 1.3 kJ/g-Pd
- H2: 1.0 kJ/g-Pd

D/Pd = 1.08

2nd phase

- D2: 1.9 kJ/g-Pd
- H2: -1.3 kJ/g-Pd

H/Pd = 1.00

Flow rate

- D: 1.76 sccm
- H: 2.29 sccm

D-PZ 3.0g-Pd
H-PZ 3.0g-Pd

S7_O5_ICCF15
Santoku Pd (PZ3,4#1)

1st phase
- D2: 2.13kJ/g-Pd
- H2: 1.70kJ/g-Pd
- D/Pd=1.07
- H/Pd=0.86

2nd phase
- D2: 1.28kJ/g-Pd
- H2: 0.26kJ/g-Pd
- D-PZ 3.0g-Pd
- H-PZ 3.0g-Pd
- Flow rate
  - D: 1.85 sccm
  - H: 2.93 sccm
Santoku Pd (PZ9,10#1)

1st phase
D₂ : 2.39 kJ/g-Pd
H₂ : 2.27 kJ/g-Pd

2nd phase
D₂ : 0.91 kJ/g-Pd
H₂ : 0.91 kJ/g-Pd

Flow rate
D : 6.42 sccm
H : 22.45 sccm

D-PZ 4.2g-Pd
H-PZ 4.2g-Pd

D/Pd = 1.41
H/Pd = 1.02

Output power [W]
Pressure [MPa]

Time [min]

Output (D₂)
Output (H₂)
Pressure (D₂)
Pressure (H₂)
<table>
<thead>
<tr>
<th>run</th>
<th>weight [g]</th>
<th>Gas</th>
<th>flow rate [sccm]</th>
<th>Output energy[kJ]</th>
<th>Specific output energy[kJ/g]</th>
<th>D/Pd or H/Pd</th>
<th>E per D/H (kJ/g) per D/H atom</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td></td>
<td></td>
<td></td>
<td>1st phase</td>
<td>2nd phase</td>
<td>1st phase</td>
<td>2nd phase</td>
</tr>
<tr>
<td>D-PP1#1</td>
<td>5</td>
<td>D2</td>
<td>3.5</td>
<td>0.5±0.4</td>
<td>2.5±4.1</td>
<td>0.10±0.07</td>
<td>0.52±0.83</td>
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<tr>
<td>D-PP1#2</td>
<td>5</td>
<td>D2</td>
<td>4.3</td>
<td>0.5±0.2</td>
<td>4.0±4.4</td>
<td>0.10±0.05</td>
<td>0.79±0.88</td>
</tr>
<tr>
<td>H-PP2#1</td>
<td>5</td>
<td>H2</td>
<td>6.8</td>
<td>0.4±0.2</td>
<td>2.6±3.9</td>
<td>0.08±0.003</td>
<td>0.53±0.8</td>
</tr>
<tr>
<td>D-PB1#1</td>
<td>3.2</td>
<td>D2</td>
<td>3.5</td>
<td>1.7±0.3</td>
<td>8.3±4.5</td>
<td>0.54±0.1</td>
<td>2.6±1.4</td>
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<td>H-PB2#1</td>
<td>3.6</td>
<td>H2</td>
<td>5.6</td>
<td>1.6±0.3</td>
<td>-2.2±4.6</td>
<td>0.45±0.08</td>
<td>-0.62±1.3</td>
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<tr>
<td>D-PB3#1</td>
<td>20</td>
<td>D2</td>
<td>2.9</td>
<td>9.3±1.1</td>
<td>1.1±0.5</td>
<td>0.47±0.06</td>
<td>0.058±0.023</td>
</tr>
<tr>
<td>D-PB3#2</td>
<td>20</td>
<td>D2</td>
<td>0.8</td>
<td>3.3±0.5</td>
<td>3.4±2.6</td>
<td>0.17±0.03</td>
<td>0.17±0.13</td>
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<tr>
<td>H-PB4#2</td>
<td>20</td>
<td>H2</td>
<td>1.9</td>
<td>3.2±0.2</td>
<td>14±4.6</td>
<td>0.16±0.01</td>
<td>0.68±0.23</td>
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<tr>
<td>H-PB4#3</td>
<td>20</td>
<td>H2</td>
<td>1.5</td>
<td>16±2.4</td>
<td>-4.8±8.1</td>
<td>0.79±0.01</td>
<td>-0.24±0.4</td>
</tr>
<tr>
<td>D-PB3#3</td>
<td>20</td>
<td>D2</td>
<td>1.1</td>
<td>14±1.7</td>
<td>-2.2±1.1</td>
<td>0.68±0.01</td>
<td>-1.1±0.54</td>
</tr>
<tr>
<td>D-PB3#4</td>
<td>20</td>
<td>D2</td>
<td>1.1</td>
<td>3.1±0.4</td>
<td>0.3±4.7</td>
<td>0.16±0.02</td>
<td>0.016±0.23</td>
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<tr>
<td>D-PZ1#1</td>
<td>10</td>
<td>D2</td>
<td>1.76</td>
<td>7.0±0.2</td>
<td>6.8±1.3</td>
<td>1.3±0.04</td>
<td>1.9±0.31</td>
</tr>
<tr>
<td>H-PZ2#1</td>
<td>10</td>
<td>H2</td>
<td>2.29</td>
<td>3.6±0.1</td>
<td>-5.1±1.4</td>
<td>1.0±0.03</td>
<td>-1.5±0.32</td>
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<tr>
<td>D-PZ3#1</td>
<td>10</td>
<td>D2</td>
<td>1.85</td>
<td>6.4±0.2</td>
<td>5.5±0.8</td>
<td>2.13±0.0</td>
<td>1.2±0.2</td>
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<tr>
<td>H-PZ4#1</td>
<td>10</td>
<td>H2</td>
<td>2.93</td>
<td>5.1±0.1</td>
<td>1.1±0.9</td>
<td>1.70±0.0</td>
<td>-1.3±0.2</td>
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<tr>
<td>D-PZ3#2</td>
<td>10</td>
<td>D2</td>
<td>1.66</td>
<td>1.7±0.03</td>
<td>9.89±1.48</td>
<td>0.03±0.070</td>
<td>2.3±0.35</td>
</tr>
<tr>
<td>H-PZ4#2</td>
<td>10</td>
<td>H2</td>
<td>2.79</td>
<td>0.58±0.05</td>
<td>1.68±1.46</td>
<td>0.17±0.011</td>
<td>0.39±0.34</td>
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<tr>
<td>D-PZ3#3</td>
<td>10</td>
<td>D2</td>
<td>1.69</td>
<td>0.29±0.04</td>
<td>-3.47±0.34</td>
<td>0.07±0.092</td>
<td>-0.81±0.35</td>
</tr>
<tr>
<td>H-PZ4#3</td>
<td>10</td>
<td>H2</td>
<td>2.99</td>
<td>0.37±0.02</td>
<td>0.75±0.35</td>
<td>0.01±0.006</td>
<td>0.17±0.34</td>
</tr>
<tr>
<td>D-PZ5#1</td>
<td>10</td>
<td>D2</td>
<td>2.02</td>
<td>7.14±0.15</td>
<td>1.26±1.36</td>
<td>2.37±0.035</td>
<td>0.29±0.32</td>
</tr>
<tr>
<td>H-PZ6#1</td>
<td>10</td>
<td>H2</td>
<td>6.23</td>
<td>7.07±0.07</td>
<td>-0.23±1.44</td>
<td>2.33±0.018</td>
<td>-0.05±0.33</td>
</tr>
<tr>
<td>D-PZ5#3</td>
<td>10</td>
<td>D2</td>
<td>9.93</td>
<td>0.54±0.025</td>
<td>0.23±1.51</td>
<td>0.18±0.008</td>
<td>0.08±0.5</td>
</tr>
<tr>
<td>H-PZ6#3</td>
<td>10</td>
<td>H2</td>
<td>10.69</td>
<td>0.92±0.025</td>
<td>4.18±1.51</td>
<td>0.31±0.008</td>
<td>1.39±0.5</td>
</tr>
<tr>
<td>D-PZ9#1</td>
<td>14</td>
<td>D2</td>
<td>6.42</td>
<td>10.23±0.10</td>
<td>3.81±1.51</td>
<td>2.44±0.024</td>
<td>0.91±0.36</td>
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<tr>
<td>H-PZ10#1</td>
<td>14</td>
<td>H2</td>
<td>22.55</td>
<td>9.56±0.034</td>
<td>3.82±1.51</td>
<td>2.28±0.008</td>
<td>0.91±0.36</td>
</tr>
</tbody>
</table>
$1^{st}$ phase results

- PP: Loading ratios are bulk values, and specific heats are also bulk values.

- PB: Loading ratios are 2-fold of bulk values, and specific heats are 3-fold of bulk values.

- PZ: Loading ratios are 2.5-fold of bulk values, and specific heats are 10-fold of bulk values.

<table>
<thead>
<tr>
<th>run</th>
<th>D/Pd or H/Pd</th>
<th>$E$ per D/H atom [eV]</th>
</tr>
</thead>
<tbody>
<tr>
<td>D-PP</td>
<td>0.46</td>
<td>0.24</td>
</tr>
<tr>
<td>H-PP</td>
<td>0.45</td>
<td>0.20</td>
</tr>
<tr>
<td>D-PB</td>
<td>0.82±0.05</td>
<td>0.67±0.02</td>
</tr>
<tr>
<td>H-PB</td>
<td>0.78</td>
<td>0.63</td>
</tr>
<tr>
<td>D-PZ</td>
<td>1.15±0.17</td>
<td>2.24±0.28</td>
</tr>
<tr>
<td>H-PZ</td>
<td>1.07±0.24</td>
<td>1.95±0.49</td>
</tr>
</tbody>
</table>

Average values:

$E$ per D/H atom [eV] for P-P: 0.76±0.05

$E$ per D/H atom [eV] for P-C: 0.62±0.03
Conclusion

• The twin system of D(H) gas loading is a useful tool.

• Nano-Palladium Zirconium-oxide composite generates 10-fold larger specific heat by D(H)-absorption, compared to that of bulk palladium.

• Nano-Palladium Zirconium-oxide composite generates excess heat in the phase-2 for D₂ gas charging.

• We need further to study dependence on flow rate, nano-particle size, and cell temperature.

• We need also study of other material samples.

• Analyses of ⁴He production and nuclear particle emission are expected.