Condensed Matter Nuclear Reactions in Nano-Materials

ARPA-E Workshop on Low-Energy Nuclear Reactions

October 21-22, 2021

With additional backup slides that weren’t presented at the Workshop. Workshop presenters with additional details are in “[...]”.

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Have We Found the Keys to the Kingdom?

- Pd used as a hydrogen gas separator/purifier. [Benyo]
  - Has a unique electronic structure: 4d^{10} <> 4d^{9}5s^{1}: \textit{paramagnetic and ferromagnetic states}
- Pd will load H/D, e.g. PdD\textsubscript{x} to x < .56, \(\alpha \gg \beta\) phase
  - Fleischmann and Pons used electrolysis to bulk load x > .9, \textit{but takes days to weeks to load}
- McKubre has shown bulk PdD\textsubscript{x}, x > .86 for onset of LENR heat. [McKubre]
- Szpak showed electrolytic Pd/D co-deposition \textit{rapidly loads} x≈ 1.0 [Mosier-Boss]
  - SEM analysis shows a size range from \textit{nm to \(\mu\)m scales}
- Miles and Barham: \textit{watts thermal}
- Storms suggests \textit{nm fissures} provide a nuclear active environment (NAE)
- Staker observed and created \textit{Pd Super-Abundant Vacancies (SAV)} allowing high-densities of hydrogen isotopes
- DeChiaro modeled \textit{itinerant ferromagnetism} and SAV
- Pianitelli, \textit{Ni}, Takahashi, Celani and others have found \textit{NiCu exhibit excess heat} with hydrogen

<table>
<thead>
<tr>
<th>Excess Heat</th>
<th>Duration</th>
<th>Mass</th>
<th>Material</th>
<th>(W_t/g)</th>
<th>Joules</th>
</tr>
</thead>
<tbody>
<tr>
<td>Arata:</td>
<td>10 (W_t)</td>
<td>12 weeks</td>
<td>3 g</td>
<td>Pd-black nanoparticles w/D\textsubscript{2}</td>
<td>3.3</td>
</tr>
<tr>
<td>Takahashi</td>
<td>226 (W_t)</td>
<td>several weeks [5]</td>
<td>505 g</td>
<td>CuNi nanoparticles w/H\textsubscript{2}</td>
<td>0.45</td>
</tr>
<tr>
<td>Ahern:</td>
<td>21 (W_t)</td>
<td>5 days</td>
<td>5 g</td>
<td>PdNi nanopowder w/D\textsubscript{2}</td>
<td>4.2</td>
</tr>
<tr>
<td>Celani:</td>
<td>18 (W_t)</td>
<td>5 hours</td>
<td>0.45 g</td>
<td>CuNi nanolayers w/H\textsubscript{2} or D\textsubscript{2}</td>
<td>40.0</td>
</tr>
</tbody>
</table>

\textit{LENR requires multiple science and engineering disciplines.}
\textit{Questions remain: Scaling, self-operating, reaction products, life-time, triggering, mechanisms.}
Common Features of LENR-Active Nano-Materials

- **Hydrogen isotope spillover**:
  - Three stages: adsorption, dissociation, absorption
  - Pd will do all three, Cu the first two, Ni absorbs H at > 200 °C

- **Materials**:
  - Pd, Pd$_3$Ni$_7$, Pd$_1$Ni$_9$, Cu$_3$Ni$_7$, Cu$_5$S$_3$I$_4$Mn$_1$ often in ZrO$_2$

- **Structures**:
  - Nano-scale structures, 2 – 20 nm

- **Fabrication**:
  - Spin-cast powders
  - Multi-layers
  - Co-deposition
  - Double cathode

- **Operation**:
  - H$_2$ or D$_2$ gas or electrolytic loading and/or flux

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Less appreciated

The roles of magnetic order and disorder

- Bulk Ni Curie Point ≈ 354 °C,
- Nanoparticle Curie Point T < bulk material
- Pd is paramagnetic
- ZrO$_2$ in contact with Pd strains the lattice induces ferromagnetism
- PdH and PdD are superconductors
- Superconductor YBCO$_x$ is LENR Active

Is LENR a topological phenomena? Is magnetism a key?

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Excess Heat

<table>
<thead>
<tr>
<th>Material</th>
<th>Excess Heat</th>
</tr>
</thead>
<tbody>
<tr>
<td>Pd, PdNi</td>
<td>$D_2$</td>
</tr>
<tr>
<td>Ni, CuNi</td>
<td>$H_2$</td>
</tr>
<tr>
<td>Oxides</td>
<td>often present</td>
</tr>
</tbody>
</table>

Comparisons

<table>
<thead>
<tr>
<th>Property</th>
<th>Value</th>
</tr>
</thead>
<tbody>
<tr>
<td>$10^4$ eV/H</td>
<td>(Iwamura)$^4$</td>
</tr>
<tr>
<td>$10^3 - 10^5$ eV/D</td>
<td>(Takahashi)$^{12}$</td>
</tr>
<tr>
<td>4.88 eV/H</td>
<td>(oxidation: H$_2$O)</td>
</tr>
</tbody>
</table>

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### Representative Nano-material Anomalous Heat

#### Arata (nano-Pd black/D₂ double cathode)¹ [Narita & Nagle]

<table>
<thead>
<tr>
<th>Material:</th>
<th>20 nm diameter, Pd black, 3 g</th>
</tr>
</thead>
<tbody>
<tr>
<td>Triggering:</td>
<td>Electrolytically loaded, [1 kbar D₂]</td>
</tr>
<tr>
<td>Excess Heat:</td>
<td>5 – 10 W, continuous</td>
</tr>
<tr>
<td>Duration:</td>
<td>12 weeks</td>
</tr>
</tbody>
</table>

#### Celani (CuNiMn & oxide nanolayers, D₂ or H₂)³

<table>
<thead>
<tr>
<th>Material:</th>
<th>Mod. Constantan [Cu₅₅Ni₄₄Mn] w/oxides, 0.45g⁴</th>
</tr>
</thead>
<tbody>
<tr>
<td>Triggering:</td>
<td>Thermal, 50 Hz A/C stimulation, 600 V, heating</td>
</tr>
<tr>
<td>Excess Heat:</td>
<td>18 Wₜ with 99.7 W Joule heating</td>
</tr>
<tr>
<td>Duration:</td>
<td>5 hours</td>
</tr>
</tbody>
</table>

#### Ahern (nano-PdNiZrO₂/D₂)²

<table>
<thead>
<tr>
<th>Material:</th>
<th>10 nm diameter PdNiZrO₂ powder, 5 g</th>
</tr>
</thead>
<tbody>
<tr>
<td>Triggering:</td>
<td>Thermal, T &gt; 360 °C. [Ni Curie Point, 358 °C]</td>
</tr>
<tr>
<td>Excess Heat:</td>
<td>~ 21 watts, not repeated</td>
</tr>
<tr>
<td>Duration:</td>
<td>5 days, terminated for evaluation</td>
</tr>
</tbody>
</table>

#### Takahashi⁵,⁶ (PdNiZrO₂ & CuNiZrO₂ nanopowders) [Narita]

<table>
<thead>
<tr>
<th>Material:</th>
<th>PdNi₁₀/ZrO₂ (PNZ10)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Cu₃Ni₇/ZrO₂ (CNZ7) 505 g</td>
</tr>
<tr>
<td>Triggering:</td>
<td>Thermal?</td>
</tr>
<tr>
<td>Excess Heat:</td>
<td>PNZ10rr 186 W/kg⁵ D₂</td>
</tr>
<tr>
<td></td>
<td>CNZ7rr 226 W/kg⁵ H₂</td>
</tr>
<tr>
<td></td>
<td>reaction energy⁵ (η-value) 10³ to 10⁵ eV/D</td>
</tr>
<tr>
<td>Duration:</td>
<td>several weeks</td>
</tr>
</tbody>
</table>

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⁴ Personal communications with Celani during and after the ARPA-E Workshop.


## Related Experiments With Excess Power

<table>
<thead>
<tr>
<th>Lead researcher/[presenter]</th>
<th>Nano-material</th>
<th>H/D Isotope</th>
<th>Country</th>
<th>Year</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ahern(^1)</td>
<td>PdNi/ZrO(_2), nanopowder</td>
<td>D(_2)</td>
<td>US</td>
<td>&lt; 2012</td>
</tr>
<tr>
<td>Arata(^2) [Narita, Nagle, McKubre]</td>
<td>Pd black nanopowder, double cathode</td>
<td>D(_2) from D(_2)O</td>
<td>Japan</td>
<td>&lt; 1997</td>
</tr>
<tr>
<td>Beiting(^3)</td>
<td>ZrO(_2),NiPd nanopowder</td>
<td>H(_2)</td>
<td>US</td>
<td>&lt; 2017</td>
</tr>
<tr>
<td>Celani(^4)</td>
<td>NiCuMn (Fe) oxide nano-layers</td>
<td>H(_2), D(_2)</td>
<td>Italy</td>
<td>&lt; 2014</td>
</tr>
<tr>
<td>Iwamura(^5) [Narita]</td>
<td>CuNi, nanolayers</td>
<td>H(_2), D(_2)</td>
<td>Japan</td>
<td>&lt; 2018</td>
</tr>
<tr>
<td>Kitamura(^6) [Narita]</td>
<td>Pd, ZrO(_2) nanopowder</td>
<td>H(_2), D(_2)</td>
<td>Japan</td>
<td>&lt; 2009</td>
</tr>
<tr>
<td>Miles(^7,8) [McKubre]</td>
<td>Pd/D Co-deposition (ammonia complex)</td>
<td>D(_2) from D(_2)O</td>
<td>US/Japan</td>
<td>&lt; 1999</td>
</tr>
<tr>
<td>Snoswell(^9)</td>
<td>Ni nanopowder</td>
<td>H(_2)?</td>
<td>Australia</td>
<td>&lt; 2012</td>
</tr>
<tr>
<td>Storms(^10) [Nagle]</td>
<td>compressed Ni or Pd micropowders</td>
<td>H(_2), D(_2)</td>
<td>US</td>
<td>&lt; 2021</td>
</tr>
<tr>
<td>Swartz(^11) [Nagle]</td>
<td>PdNi-ZrO(_2), PdZrO(_2) nanopowder</td>
<td>D(_2)</td>
<td>US</td>
<td>&lt; 2017</td>
</tr>
<tr>
<td>Takahashi(^12) [Narita]</td>
<td>PdNi-ZrO(_2) and CuNi-ZrO(_2) nanopowder</td>
<td>H(_2), D(_2)</td>
<td>Japan</td>
<td>&lt; 2013</td>
</tr>
</tbody>
</table>

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9. Communication between B. Ahern and M. Snoswell, discussing Quantum Sphere Corp. nano-nickel powders. See *EPRI Report 1025575*, p. 4-7, ref [30].


Assessment of Needs

- **Experimental Improvement** (*minimum sensitivity to detect rare products or swamped by background*)
  - High Temperature Calorimetry
    - Range 30 C – 500 C, > 100 mW sensitivity
  - Nuclear, HPGe, RF, IR, UV diagnostics, (> 5 sigma over background)
    - *in situ and adjacent to operating devices*: watch out for cosmogenic muons and neutrons, natural BG radiation
    - Real-time energetic particle spectroscopy preferred: TOF if possible (gold standard for particle energy)
    - Witness materials with HPGe in situ, post HPGe monitoring and material assays
- **Material assay and limitations (interferences)** Indicates the roles of various structures and materials, contaminants
  - SEM/EDX: wide area FOV, qualitative surface structure, limited elemental sensitivity (parts per ten thousand)
  - ICP-MS: quantitative, isotope specific, but multi-ion (e.g. $H_2^+$, Ar feed gas) complexes, ppb-ppm
  - TOF-SIMS: isotope specific, qualitative, small FOV, ppb-ppm
  - NAA/PGNAA: quantitative, isotope specific, limited isotope detection, ppt-ppm
  - Alpha/Beta Liquid Scintillator Spectrometer: tritium, activation products, limited energy resolution, ppm

- **Patent Rights**
  - USPTO treats LENR heat as “perpetual motion”, hence, unpatentable, yet necessary for investment and commercialization

- **Licensing**
  - NRC and IAEA providence
  - Patents and patent interference

- **Health Physics Concerns**
  - Nanoparticles, especially non-encapsulated Ni-based nanopowder and their escape via seal and valve seat fouling
  - Self-generated B and EM fields
  - Bremsstrahlung radiation
  - Neutron radiation

*Fundamentally, there is a need to run longer with simultaneous diagnostic measurements to correlate effects.*
Preferred Experiments

**• Diagnostics:**

**Nuclear:** HPGe x-ray/γ 10 keV – 3 MeV or LaBr3 or NaI(Tl) [in decreasing energy resolution, but increasing detection (size) efficiency]

CR-39, in situ (< 50 C), external [Detect both charged particles and neutrons, speciation and energy resolution]

Neutron scintillators for time resolved (< 200 nsec resolution) [neutron counting and/or spectroscopy]

Plastic/glass scintillators *(in situ ion counting)*, (< 20 nsec resolution) [Detect charged particles]

Silicon Barrier detectors (200 keV – 20+ MeV ion detection) [Resolve charged particle energy and nuclear exit channel]

**Optical:** Fiberoptic feedthrough for spectroscopy, digital camera [Detect spectra and spatial relationship of “hot” spots]

IR: NIR, MIR 1 um – 12 um range, notch-filters [Detect down-shifted bremsstrahlung, non-black-body radiation]

RF: 100 kHz – 10 GHz, with H/V Stokes parameter polarization [Detect magnetic effects]

**Low temperature Calorimetry:** 25 C – 100+ °C, < 50 mWₜ sensitivity [suitable to aqueous electrolysis]

**High temperature Calorimetry:** 100 C – 500+ °C, 200 mWₜ sensitivity [suitable to high temp gas operation]

**• Experiments:**

Electrolytic co-deposition with fractal surface nano-particles:

PdCl₂, LiCl electrolyte with Pt, Ag, Au cathodes, e.g. patent¹ and Trackers STEM Protocol™

Operation > 100 °C, approaching 150 C, 15 bar [determine effect lifetime by running longer]

Slow, ramped up V/I for 2+ weeks

Fast, constant or ramped down, V/I, for 2 weeks [determine effect lifetime by running longer: Letts/Dahlgren/HIVER]

Gas Cycling of nanopowders and nanolayers:

nanopowders and nanolayers, H₂, D₂, HD, He, Ar

Operation > 200 °C, << 1 bar (often less than 100 torr)

Double Cathode with nano-powders:

nano-powders placed within sealed, electrolyzed Pd tubing for high loading

**• Triggers:**

**Low Energy:** thermal, RF, laser, gas cycling/flux

**High Energy:** neutron, e-beam, bremsstrahlung

Possible ARPA-E Program Going Forward

- Drive towards LENR/Lattice Confinement Fusion (LCF) Scaling
  - Self-sustaining operation
    - Establish LENR/LCF lifetime, power output/gm, available Delta T
    - Thermal or direct power conversion
  - Path to Commercialization: Watts, kW or MW?
  - Investigate multiple LENR/LCF-capable experiments: “Lab rats”
  - Develop predictive modeling of materials and nuclear effects
  - Setup cooperative teams: share knowledge and findings!
  - Publish results in “Tier 1” journals
    - Recognition by scientific and engineering communities
    - Acceptance by government agencies and universities
    - Harness private sector funding

- Beyond ARPA-E Program
  - Multi-agency supported LENR/LCF Program
  - Foreign entity participation
  - Develop university and student support to create the technology workforce
  - Deploy the technology

Catch-22: Government agencies won’t recognize LENR unless Tier 1 journals publish results. These journals won’t publish results unless LENR is recognized by these agencies.¹

Backup Slides

Backup material wasn’t presented at the ARPA-E Workshop. However, both Takahashi’s and Iwamura’s research were discussed by Narita in the Workshop.
Takahashi¹,²: Nano-particle Anomalous Heat (AHE)

Kobe University C system AHE calorimetry Schematic, oil-flow-calorimeter system with flow-rate-monitors and dual heaters (W1,W2).

Observations

PdNi₁₀/ZrO₂ (PNZ10) Cu₃Ni₇/ZrO₂ (CNZ7) melt-spun, calcinated or recalcinated nano-powders.

Calorimetry¹

80–400 Wₑₓ/kg excess thermal repeatable for several weeks at elevated temperature ≈ 300 °C with re-calcined PNZ-type with D.

Specific reaction energy² (η-value) 10³ to 10⁵ eV/D.

50–140 Wₑₓ/kg obtained by CNZ-type with H

50–70 Wₑₓ > two weeks by 505 g CNZ7r (re-calcined)

Neutron Counting¹

Neutron yield: 0.1 n/J: 1.0 × 10⁻¹³ of bare d–d fusion.

AHE is largely aneutronic!

Iwamura¹: Nano-metal Composite Anomalous Heat

**Experiment**

- **Six-layer nano-composite**

<table>
<thead>
<tr>
<th>Ni 14 nm</th>
<th>Cu 2 nm</th>
</tr>
</thead>
<tbody>
<tr>
<td>Ni 14 nm</td>
<td>Cu 2 nm</td>
</tr>
<tr>
<td>Ni 14 nm</td>
<td>Cu 2 nm</td>
</tr>
<tr>
<td>Ni 14 nm</td>
<td>Cu 2 nm</td>
</tr>
<tr>
<td>Ni 14 nm</td>
<td>Cu 2 nm</td>
</tr>
<tr>
<td>Ni 14 nm</td>
<td>Cu 2 nm</td>
</tr>
<tr>
<td>Ni 0.1 mm</td>
<td>Cu 2 nm</td>
</tr>
</tbody>
</table>

**Released excess energy per hydrogen**

<table>
<thead>
<tr>
<th></th>
<th>Six Layers (Cu 2nm, Ni 14nm)</th>
</tr>
</thead>
<tbody>
<tr>
<td></td>
<td>Total Excess: $1.1 \times 10^6$ J</td>
</tr>
<tr>
<td>Absorbed H [mol]</td>
<td>$7.3 \times 10^{-4}$</td>
</tr>
<tr>
<td>Absorbed number of H</td>
<td>$4.4 \times 10^{20}$</td>
</tr>
<tr>
<td>Total excess energy [J]</td>
<td>$1.1 \times 10^6$</td>
</tr>
<tr>
<td>Excess energy per hydrogen [J/H-mol]</td>
<td>$1.5 \times 10^9$</td>
</tr>
<tr>
<td>Excess energy per hydrogen [eV/H]</td>
<td>$1.6 \times 10^4$</td>
</tr>
</tbody>
</table>

**Comparisons**

- $10^4$ eV/H (Iwamura) vs $10^3 - 10^5$ eV/D (Takahashi) vs $4.88$ eV/H $> \text{H}_2\text{O}$ (oxidation)

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Nano-material Anomalous Heat

Ahern (2011, nanopowders) ¹

"...nanotextured nickel in the presence of hydrogen gas, tests of similar materials conducted under this EPRI research grant produced only milliwatt-scale thermal power releases... one experiment, a 21-watt release was observed but not replicated."

Reproducibility depended upon triggering by raising temperature above 360 °C. The excess heating rate output did not exceed 200 milliwatts.

...thermal power estimated to be 100 milliwatts per degree C of elevation above the value of the outer resistance thermal device (RTD)....10-nm nickel powder from Quantum Sphere Corp. The inner RTD was 208°C hotter than the outer RTD (533 °C versus 325 °C) ...~ 21 watts from 5 grams of nanopowder... maintained for five days... terminated for evaluation.

**Issue**: Is heat produced by global collapse of magnetic order at Curie Point or are reactions initiated by the same? *It appears that the excess power over time exceeds the magnetic field collapse energy storage.*

Celani (< 2020, nanolayers)²

...the successful work of Brian Ahern with nickel-based powders and having close experience with similar systems, we speculated on the possibility of an unidentified “trigger”... AHE can be triggered and controlled electrically, opening the possibility for further scale up toward practical applications...we achieve 8–9W of AHE when using 80W of Joule heating of the Constantan [Cu₃₅Ni₄₄Mn₁] under AC stimulation (50 Hz, up to 600V).

Constantan wires producing the largest AHE were produced in the 1970s which contained about 0.5% iron, nearly absent in more recent Constantan lots. Holmlid reported the possible formation of ultradense states of atomic hydrogen and deuterium in iron-potassium catalysts.

**Takahashi**³⁴ (nanopowders) [see Narita overview of Japanese research]

PdNi₁₀/ZrO₂ (PNZ10) Cu₃₅Ni₇/ZrO₂ (CNZ7) melt-spun, calcinated or recalcinated nanopowders. 80–400 W/kg excess thermal Wₓ, repeatable for several weeks at elevated temperature ≈ 300 °C with re-calcined PNZ-type with D³. Specific reaction energy⁴ (η-value) 10³ to 10⁶ eV/D. 50-140 W/kg Wₓ, obtained by CNZ-type with H and 50–70 W continued > two weeks by 505 g CNZ7r (re-calcined)

Neutron yield: 0.1 n/J: 1.0 × 10⁻¹³ of bare d–d fusion. *AHE is largely aneutronic!*

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