CR-39 Results Obtained Using Pd/D Co-deposition

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Lawrence P.G. Forsley
SRI Replication of CR-39 Results
60 µm PE between CR-39 & Ag/Pd/D cathode

LET curves indicate that 60 µm PE will block 7 MeV alphas and 1.8 MeV protons

The detector underwent microscopic examination, it was scanned, and sequentially etched
Microscopic Analysis SRI Detector

60 µm PE film between cathode and detector
Two Triple Tracks were Observed on the SRI detectors: Evidence of > 9.6 MeV Neutrons

Johan Frenje, MIT, “I must say that the data and their analysis seem to suggest that energetic neutrons have been produced,” (ACS, 2009)
Ohmic measurements indicated that the Pd metal had not gone through the PE film

Tracks correlated with the Pd deposit

- Pd deposit is the source of the tracks
Example of a Scanned Image

image

objects identified

focus inside pits

green = tracks
Automated Scanner Results Obtained for the CR-39 Detector used in the SRI Replication
Sequential Etching Analysis (Lipson and Roussetski)

Track density, [cm$^{-2}$]

Track diameter, [µm]

proton recoil energy (MeV)

\begin{itemize}
  \item 0.5
  \item 1.3
  \item 2.1
  \item 2.9
  \item 3.7
  \item 4.5
\end{itemize}

\begin{itemize}
  \item 2.45 MeV (DD) Neutrons
  \item Pd/D co-dep
  \item $^{252}$Cf neutron source
\end{itemize}

CR-39 outside cell, 14 hr etch

detector inside cell, 21 hour etch

LET Spectrum Analysis (Zhou, NASA)

\begin{itemize}
  \item FRONT SIDE
    \begin{itemize}
      \item proton, 10-5
      \item proton, 10-6
      \item alpha, 10-5
      \item alpha, 10-6
    \end{itemize}
    \begin{itemize}
      \item Number of Particles
      \item Energy (MeV)
    \end{itemize}
  \item BACK SIDE
    \begin{itemize}
      \item proton, 10-5
      \item proton, 10-6
      \item alpha, 10-5
      \item alpha, 10-6
    \end{itemize}
    \begin{itemize}
      \item Number of Particles
      \item Energy (MeV)
    \end{itemize}
\end{itemize}
The 3.4-14 MeV protons are 12.6-17.5 MeV p that have been slowed down by the Pd, water film, and PE film.

Expect a continuum of energies. But there is a trough at ~11 MeV.

This trough suggests that protons with these energies are being consumed.

\[ \text{natPd} + p \rightarrow 105\text{Ag} \]
\[ \text{natPd} + p \rightarrow 106m\text{Ag} \]

<table>
<thead>
<tr>
<th>Nuclide</th>
<th>Pd, p Reaction</th>
<th>Half-Life</th>
<th>Decay Mode</th>
<th>Daughter Isotope</th>
</tr>
</thead>
<tbody>
<tr>
<td>105Ag</td>
<td>105Pd(p,n)\text{105Ag}</td>
<td>41.29 d</td>
<td>(\beta^+)</td>
<td>\text{105Pd}</td>
</tr>
<tr>
<td></td>
<td>106Pd(p,2n)\text{105Ag}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>108Pd(p,4n)\text{105Ag}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td></td>
<td>110Pd(p,6n)\text{105Ag}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>105mAg</td>
<td>Same as for 105Ag</td>
<td>7.23 min</td>
<td>IT (99.66%)</td>
<td>105Ag, 105Pd</td>
</tr>
<tr>
<td></td>
<td>(\beta^+) (0.34%)</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>106mAg</td>
<td>106Pd(p,n)\text{106mAg}</td>
<td>8.28 d</td>
<td>(\beta^+)</td>
<td>106Pd, 106Ag</td>
</tr>
<tr>
<td></td>
<td>108Pd(p,3n)\text{106mAg}</td>
<td></td>
<td>IT (4.16x10^{-6}%)</td>
<td></td>
</tr>
<tr>
<td></td>
<td>110Pd(p,5n)\text{106mAg}</td>
<td></td>
<td></td>
<td></td>
</tr>
<tr>
<td>110mAg</td>
<td>110Pd(p,n)\text{110mAg}</td>
<td>249.8 d</td>
<td>(\beta^-) (98.64 %)</td>
<td>110Cd, 110Ag</td>
</tr>
<tr>
<td></td>
<td></td>
<td></td>
<td>IT (1.36 %)</td>
<td></td>
</tr>
</tbody>
</table>

Will see Ag that decays back to Pd

Will see Ag that decays to Cd
Silver was observed in high, localized concentrations shortly after electrolysis. Examination 15 months later showed the presence of cadmium in addition to silver. Changes in ratio between Ag $L_{\beta_1}$ and Ag $L_{\alpha_1}$ peak indicated that Ag is slowly changing to Cd.

- The Ag $L_{\beta_1}$ peak overlaps with the Cd $L_{\alpha}$ peaks.
Review of Analysis of SRI Detectors

- **Microscopic analysis and Automated Analysis (major/minor axis analysis)**
  - Neutrons: 2.5 MeV and > 12 MeV
  - Charged particles: > 10 MeV protons, energetic alphas

- **Sequential Etching**
  - Neutrons: 2.5 MeV
  - Charged particles: 3 MeV $p^+$, 12 MeV and 16 MeV alphas

- **Linear Energy Transfer Function Analysis**
  - Protons: 2.5 – 15 MeV
  - Alphas: Continuum of alpha energies, possible neutron recoils

Three methods of analysis yielded complementary results
The observed protons and neutrons can be accounted for by the following primary (1 and 2) and secondary (3 and 4) fusion reactions:

\[ D + D \rightarrow T (1.01 \text{ MeV}) + p (3.02 \text{ MeV}) \quad (1) \]
\[ D + D \rightarrow n (2.45 \text{ MeV}) + ^3\text{He} (0.82 \text{ MeV}) \quad (2) \]
\[ D + T (\leq 1.01 \text{ MeV}) \rightarrow \alpha (6.7-1.4 \text{ MeV}) + n (11.9-17.2 \text{ MeV}) \quad (3) \]
\[ D + ^3\text{He} (\leq 0.82 \text{ MeV}) \rightarrow \alpha (6.6-1.7 \text{ MeV}) + p (12.6-17.5 \text{ MeV}) \quad (4) \]
Effect of 60 µm PE Film

No PE Film

PE film

PE film blocks < 7 MeV α, 0.82 MeV $^3$He, and 1.01 MeV T
Zhou indicated that the effect 60 µm PE film will have on the energies of the charged particles was taken into account.

LET curves indicate that:

- > 11 MeV protons will traverse through the 1 mm thick CR-39 detector and PE film
- 60 µm PE will block 7 MeV alphas
From Zhou’s Analysis (of Both Detectors):

- The alpha and 0-9 MeV protons tracks (643) on the backside are actually due to neutrons ($D + D \rightarrow ^3He + n$)

- Frontside alpha tracks (18200) are due to long range alphas (LRA)
  - The 1-7 MeV alphas are due to 7-15 MeV alphas that have been slowed down by the Pd, water film, and PE film

- Frontside $p$ tracks (9873) between 2.6-3.4 MeV due to $p$ ($D + D \rightarrow p + t$)

- Frontside $p$ tracks (51734) between 3.4-15 MeV due to $p$ ($D + ^3He \rightarrow \alpha + p$ (12.6-17.5 MeV))
  - The 3.4-12 MeV protons are 12.6-17.5 MeV $p$ that have been slowed down by the Pd, water film, and PE film
# Primary Reaction Branching Ratio: Estimated Number of DD Neutrons (10^{-5} & 10^{-6})

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Products</th>
<th>Tracks</th>
</tr>
</thead>
<tbody>
<tr>
<td>$^{2}D + ^{2}D \rightarrow ^{3}He (0.82 \text{ MeV}) + \text{ blocked}$</td>
<td></td>
<td>643 tracks (back)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>Corrected # tracks = 1286 (front &amp; back)</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\epsilon^* = 1.17 \times 10^{-4}$</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$n = 1.1 \times 10^7$</td>
</tr>
</tbody>
</table>


‡ Confirmation: analysis of CR-39 used in Mylar experiment

- 248 DD n tracks
- $\epsilon = 1.17 \times 10^{-4}$
- 30% of tracks are elliptical
- $n = 3.03 \times 10^6$ (for one detector)
- $n = 6.06 \times 10^6$ (for two detectors)

![Proton recoil energy distribution](attachment:image.png)
Primary Reaction Branching Ratio: Estimated Number of DD Protons (10^-5 & 10^-6)

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Products</th>
<th>Number of Tracks</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D + D \rightarrow$</td>
<td>$^3He$ (0.82 MeV) + blocked</td>
<td>643 tracks (back); Corrected # tracks = 1286 (front &amp; back)</td>
</tr>
<tr>
<td>$D + D \rightarrow$</td>
<td>$T$ (1.01 MeV) + blocked</td>
<td>9873 tracks; Corrected # tracks = 19746</td>
</tr>
</tbody>
</table>

- $p (3.02$ MeV) $\geq 1.32 \times 10^6$
- $n (2.45$ MeV) $\geq 1.1 \times 10^7$
- $\epsilon = 1.17 \times 10^{-4}$
- $n/p$ branching ratio is 8.3. This is the maximum value of the $n/p$ branching ratio as the number $p$ of protons is underestimated.

- Approximately half of the tracks were counted by the scanner.
- Need to take into account the absorption of charged particles during their escape from the bulk of a thick sample, whose thickness is several times greater than the stopping range of 3 MeV protons in Pd – use TRIM (Transport of Ions in Matter).
- Most of the protons traveling through 15 µm of Pd will reach the detector.
- The Ag/Pd layer is ~ 1 mm thick.
- Number of protons is off by a factor of ~66.67.

Estimated n/p branching ratio is 8.3. This is the maximum value of the n/p branching ratio as the number p of protons is underestimated.
Used 40-60 µm thick Au/Pd/PdO heterostructures that were electrochemically loaded

\[ I_n = (19 \pm 2) \times 10^{-3} \text{ n/s} \quad \text{and} \quad I_p = (4.0 \pm 1.0) \times 10^{-3} \text{ p/s} \] 

in a 4\pi solid angle

- The lower level of proton emissions is attributed to the absorption of charged particles during their escape from the bulk of a thick sample, whose thickness is several times greater than the stopping range of 3 MeV protons in Pd

\[ \frac{n}{p} \text{ ratio estimated to be 4.75} \]
### Secondary Reaction Branching Ratio:

**Estimated Number of DT Neutrons (10^-5 & 10^-6)**

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Products</th>
<th>Neutrons (11.9-17.2 MeV)</th>
<th>P (12.6-17.5 MeV)</th>
</tr>
</thead>
<tbody>
<tr>
<td>D + T →</td>
<td>α (6.7 - 1.4 MeV) + blocked</td>
<td></td>
<td></td>
</tr>
<tr>
<td>D + $^3$He →</td>
<td>α (6.6 - 1.7 MeV) + blocked</td>
<td></td>
<td></td>
</tr>
</tbody>
</table>

2 triple tracks  
$\varepsilon_{DT} = 5.0 \times 10^{-5}$, $n = 1.18 \times 10^6$


3.38% of the DT generated tracks were triple tracks

e$\varepsilon_{DT} = 5.0 \times 10^{-5}$ is for all three types of interactions
### Secondary Reaction Branching Ratio:
#### Estimated Number of D³He Protons (10-5 &10-6)

<table>
<thead>
<tr>
<th>Reaction</th>
<th>Product</th>
<th>Energy (MeV)</th>
<th>Blocking</th>
</tr>
</thead>
<tbody>
<tr>
<td>$D + T \rightarrow$</td>
<td>$\alpha (6.7-1.4 \text{ MeV}) +$ blocked</td>
<td>n (11.9-17.2 MeV)</td>
<td>2 triple tracks</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$\varepsilon_{DT} = 5.0 \times 10^{-5}$, n = 1.18 $\times 10^6$</td>
<td></td>
</tr>
<tr>
<td>$D + ^3\text{He} \rightarrow$</td>
<td>$\alpha (6.6-1.7 \text{ MeV}) +$ blocked</td>
<td>p (12.6-17.5 MeV)</td>
<td>51734 tracks, Corrected # tracks = 103468</td>
</tr>
<tr>
<td></td>
<td></td>
<td>$p = 2.83 \times 10^5$ to 3.28 $\times 10^5$</td>
<td></td>
</tr>
</tbody>
</table>

- Approximately half of the tracks were counted by the scanner
- TRIM calculations:
  - 12.6 MeV protons traveling through 315 µm of Pd will reach the detector. The Ag/Pd layer is ~1 mm thick. Number of protons is off by a factor of ~3.17
  - Most of the 17.5 MeV protons traveling through 365 µm of Pd will reach the detector. The Ag/Pd layer is ~1 mm thick. Number of protons is off by a factor of ~2.74
## Summary on Branching Ratios

<table>
<thead>
<tr>
<th>Reagents</th>
<th>Reaction Products</th>
</tr>
</thead>
<tbody>
<tr>
<td>D + D</td>
<td>T (1.01 MeV)</td>
</tr>
<tr>
<td></td>
<td># of tritons &gt; 1.32 x 10⁶</td>
</tr>
<tr>
<td></td>
<td>p (3.02 MeV)</td>
</tr>
<tr>
<td></td>
<td># of protons &gt; 1.32 x 10⁶</td>
</tr>
<tr>
<td>D + D</td>
<td>³He (0.82 MeV)</td>
</tr>
<tr>
<td></td>
<td># of ³He = 1.1 x 10⁷</td>
</tr>
<tr>
<td></td>
<td>n (2.45 MeV)</td>
</tr>
<tr>
<td></td>
<td># of neutrons = 1.1 x 10⁷</td>
</tr>
<tr>
<td>D + T</td>
<td>α (6.7-1.4 MeV)</td>
</tr>
<tr>
<td></td>
<td># of alphas = 1.18 x 10⁶</td>
</tr>
<tr>
<td></td>
<td>n (11.9-17.5 MeV)</td>
</tr>
<tr>
<td></td>
<td># of neutrons = 1.18 x 10⁶</td>
</tr>
<tr>
<td>D + ³He</td>
<td>α (6.6-1.7 MeV)</td>
</tr>
<tr>
<td></td>
<td># of alphas = 2.83x10⁵ to 3.28x10⁵</td>
</tr>
<tr>
<td></td>
<td>p (12.6-17.5 MeV)</td>
</tr>
<tr>
<td></td>
<td># of protons = 2.83x10⁵ to 3.28x10⁵</td>
</tr>
</tbody>
</table>

- Indicates that the primary reactions are approximately equal
- Indicates that DT reactions are slightly favored over ³HeD reactions
### Efficiency of Secondary Reactions

<table>
<thead>
<tr>
<th>Reaction</th>
<th>$\sigma$ at 10 keV (barn)</th>
<th>$\sigma$ at 100 keV (barn)</th>
<th>$\sigma_{max}$ (barn)</th>
</tr>
</thead>
<tbody>
<tr>
<td>$\text{D} + \text{D} \rightarrow \text{T} + \text{p}$</td>
<td>$2.81 \times 10^{-4}$</td>
<td>$3.3 \times 10^{-2}$</td>
<td>0.096</td>
</tr>
<tr>
<td>$\text{D} + \text{D} \rightarrow \text{He} + \text{n}$</td>
<td>$2.78 \times 10^{-4}$</td>
<td>$3.7 \times 10^{-2}$</td>
<td>0.11</td>
</tr>
<tr>
<td>$\text{D} + \text{T} \rightarrow \text{\alpha} + \text{n}$</td>
<td>$2.72 \times 10^{-2}$</td>
<td>3.43</td>
<td>5.0</td>
</tr>
<tr>
<td>$\text{D} + \text{He} \rightarrow \text{\alpha} + \text{p}$</td>
<td>$2.2 \times 10^{-7}$</td>
<td>0.1</td>
<td>0.9</td>
</tr>
</tbody>
</table>

Expect more DT reactions than D$^3$He reactions
## Summary on Branching Ratios

<table>
<thead>
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<tbody>
<tr>
<td>D + D</td>
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<td># of tritons &gt; 1.32 x 10^6</td>
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<tr>
<td></td>
<td>p (3.02 MeV)</td>
</tr>
<tr>
<td></td>
<td># of protons &gt; 1.32 x 10^6</td>
</tr>
<tr>
<td>D + D</td>
<td>^3He (0.82 MeV)</td>
</tr>
<tr>
<td></td>
<td># of ^3He = 1.1 x 10^7</td>
</tr>
<tr>
<td></td>
<td>n (2.45 MeV)</td>
</tr>
<tr>
<td></td>
<td># of neutrons = 1.1 x 10^7</td>
</tr>
<tr>
<td>D + T</td>
<td>α (6.7-1.4 MeV)</td>
</tr>
<tr>
<td></td>
<td># of alphas = 1.18 x 10^6</td>
</tr>
<tr>
<td></td>
<td>n (11.9-17.5 MeV)</td>
</tr>
<tr>
<td></td>
<td># of neutrons = 1.18 x 10^6</td>
</tr>
<tr>
<td>D + ^3He</td>
<td>α (6.6-1.7 MeV)</td>
</tr>
<tr>
<td></td>
<td># of alphas = 1.80 x 10^5 to 3.18 x 10^5</td>
</tr>
<tr>
<td></td>
<td>p (12.6-17.5 MeV)</td>
</tr>
<tr>
<td></td>
<td># of protons = 1.80 x 10^5 to 3.18 x 10^5</td>
</tr>
</tbody>
</table>

- Indicates that most of the tritons produced are consumed to create 11.9-17.5 MeV neutrons
  - Secondary reactions have a higher cross section and occur at lower energies compared to primary reactions
  - A 1.01 MeV triton, once born, can go through 4.12 µm Pd – equivalent to passing through 10,236 unit cells in the lattice
  - Bockris has reported seeing a loss of tritium during Pd/D co-deposition
Bockris: Tritium in Pd/D Co-deposition
ICCF3 (1992)

▼ Dashed lines indicate the calculated expected concentrations of tritium in the solution and gas phases.
▼ Pd/D co-dep on Au: 6 out of 9 experiments showed tritium production
▼ Tritium production was observed when low tritiated D$_2$O was used.
  ▪ A burst of tritium was observed in the gas phase. At the same time, or with a slight delay, a burst of tritium occurred in the solution phase.
  ▪ A loss of tritium was observed in the solution phase when high tritiated D$_2$O was used. Suggests that the tritium is being consumed
▼ At ICCF17, Koreans reported similar results using closed cells
In all the experiments the newly found elements or isotopes appear to be explainable through occurrence of multiple deuteron captures in one or more of the isotopes of the high Z elements in/on the cathode, followed by fission of some of the complex intermediate compound nuclei.
Transmutation

Au/Pd-D, external B field

• Lorentz forces cause the deposit to form star-like features
• EDX shows a small Pd peak and the presence of Fe, Cr, Ni, and Al
  — EDX detection limits are on the order of 0.1%
  — Distribution on new elements is inhomogeneous
  — These same elements have been reported by others using a wide variety of conditions
• Are the new elements the result of multi-body deuteron fusion or the disintegration of the Pd lattice?
  — The relative size of the Pd peak suggests the latter
Different Spots on the Same Cathode
Fission reactions produce 7-16 MeV alphas (long range alphas).
As the source of the long range alphas is fission, it is very likely that the new elements observed in the EDX spectrum result from fissioning of Pd.
Conclusions

• CR-39 detectors, used in Pd/D co-deposition experiments, were subjected to microscopic analysis, automated scanning, sequential etching, and LET spectrum analysis to identify the particles responsible for the tracks
  – Particles identified were 2.45 MeV neutrons, 3-10 MeV protons, 2-15 MeV alphas, and 14.1 MeV neutrons

• Nature of the nuclear reactions
  – Protons, neutrons, and 2-7 MeV alphas observed in CR-39 detectors used in Pd/D co-deposition have energies consistent with those obtained from primary and secondary fusion reactions
  – Branching ratio of primary reactions is close to unity
  – DT reactions are favored over $^3$HeD reactions
  – Transmutation is probably the result of fissioning of the Pd nucleus. This is supported by the observation of long range alphas (7-15 MeV)
ACKNOWLEDGEMENTS

- Mitchell Swartz, Gayle Verner, and Peter Hagelstein for organizing the colloquium
- Peter for asking hard questions
- Frank Gordon, Larry Forsley, and Dr. Khim for their support
- Stan Szpak for developing the Pd/D co-deposition protocol
- Fran Tanzella and Ben Earle for doing the replication
- My husband, Roger Boss, and kids (Matt, Nathan, & Jacob) for putting up with years of revolving schedules around my experiments