

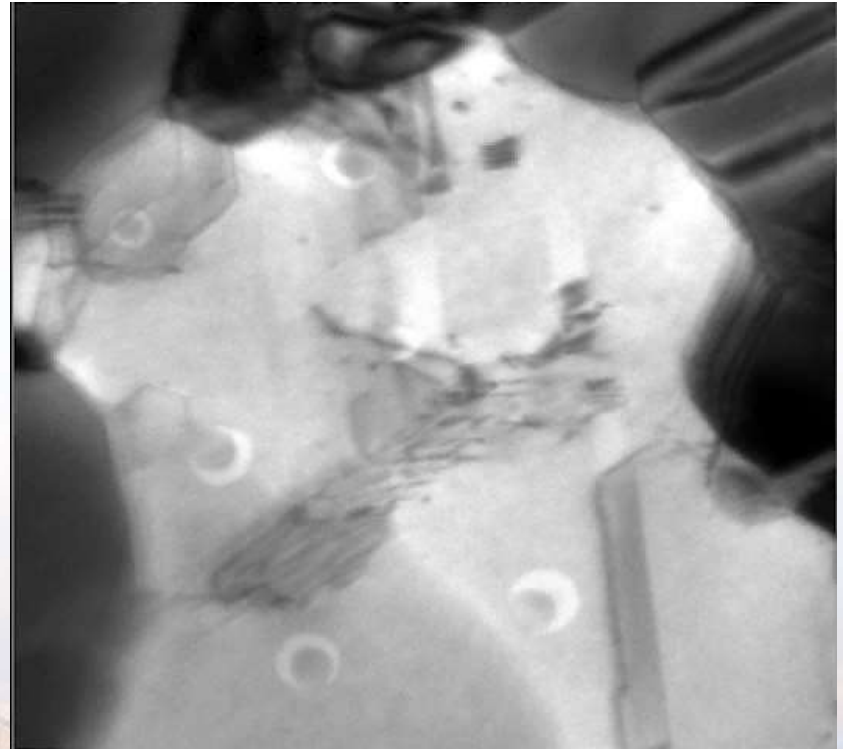
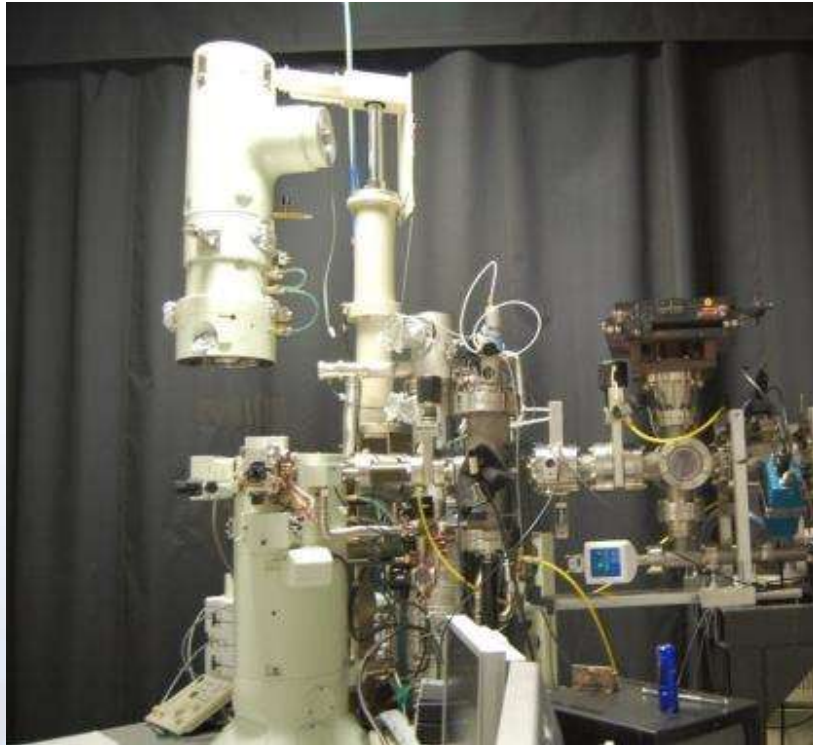
Recent Developments in Sandia's *In situ* Ion Irradiation TEM

SAND2013-6570C

K. Hattar, S.H. Pratt, C. Chisholm, O.J. El-Atwani, B.A. Hernandez-Sanchez, H. Bei, E.P. George, P. Hosemann, & A.M. Minor

Sandia's Ion Beam Lab

August 7, 2013



Sandia's *In situ* Ion Irradiation TEM is operational, but still in development



Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.

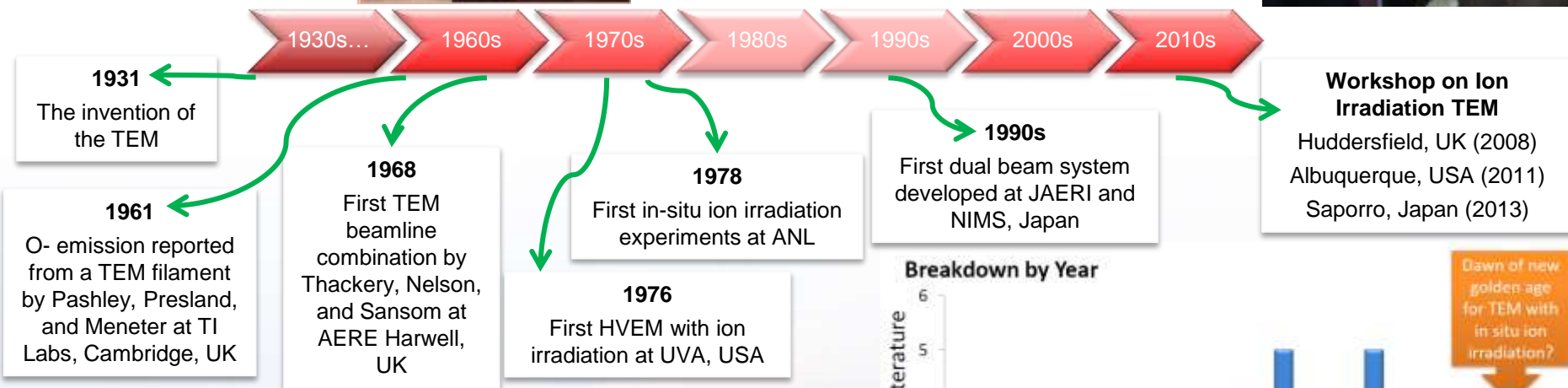


Sandia National Laboratories

History of *In situ* Ion Irradiation TEM



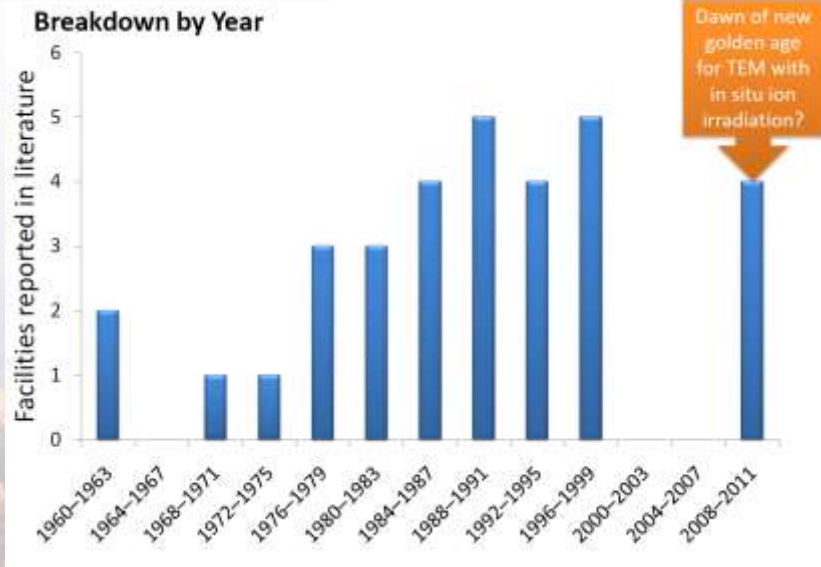
Courtesy of: J. Hinks



“The direct observation of ion damage in the electron microscope thus represents a powerful means of studying radiation damage”



D.W. Pashley and A.E.B. Presland Phil Mag. 6(68) 1961 p. 1003

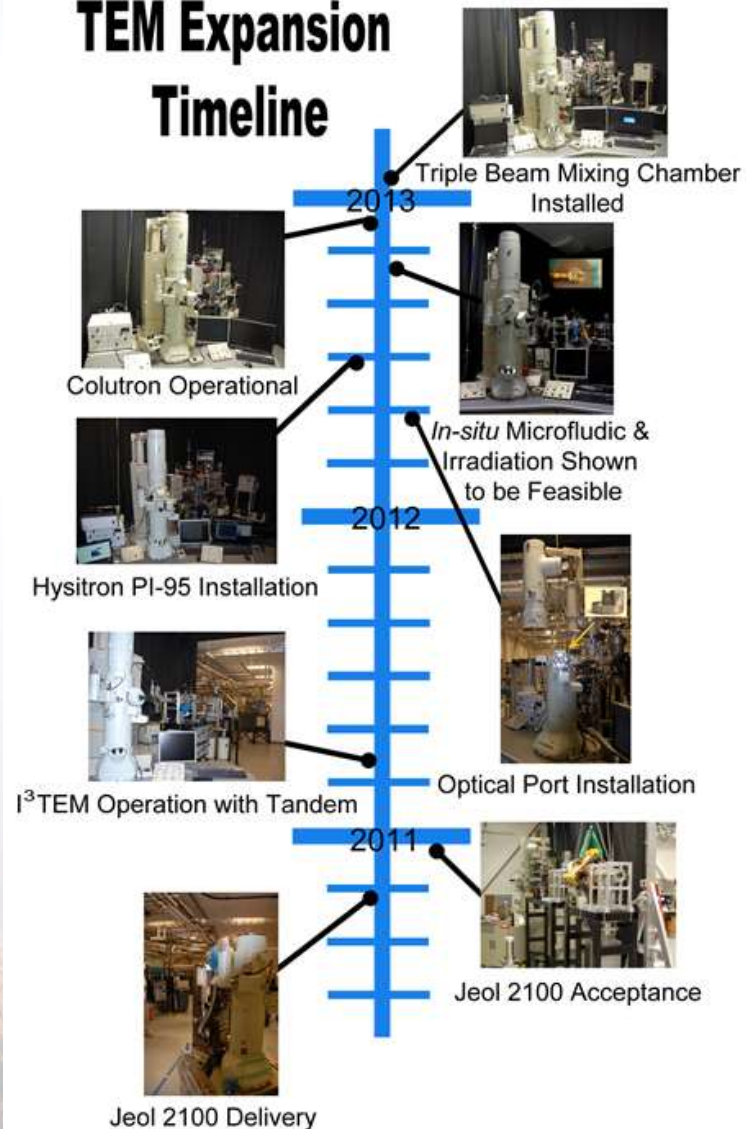


The Development of an *In situ* Ion Irradiation TEM in Sandia's New Ion Beam Lab



A special 72 wheeled vehicle with independent steering for each pair of wheels was used to move the Tandem accelerator

TEM Expansion Timeline



New Facility
laboratory space
1850 m²
office space
650 m²
Old Facility:
1300 m² total

Building: \$20M
Equipment: \$11M
Total: \$40M

Current Status of the *In situ* TEM Beamline

Collaborators: D.L. Buller & J.A. Scott



Beam burn from
14 MeV Si

Gas Heating TEM
Stage Controls

Microfluidic TEM
Stage Controls

Double tilt stage
needs to tilt only 12°

10 kV
Colutron

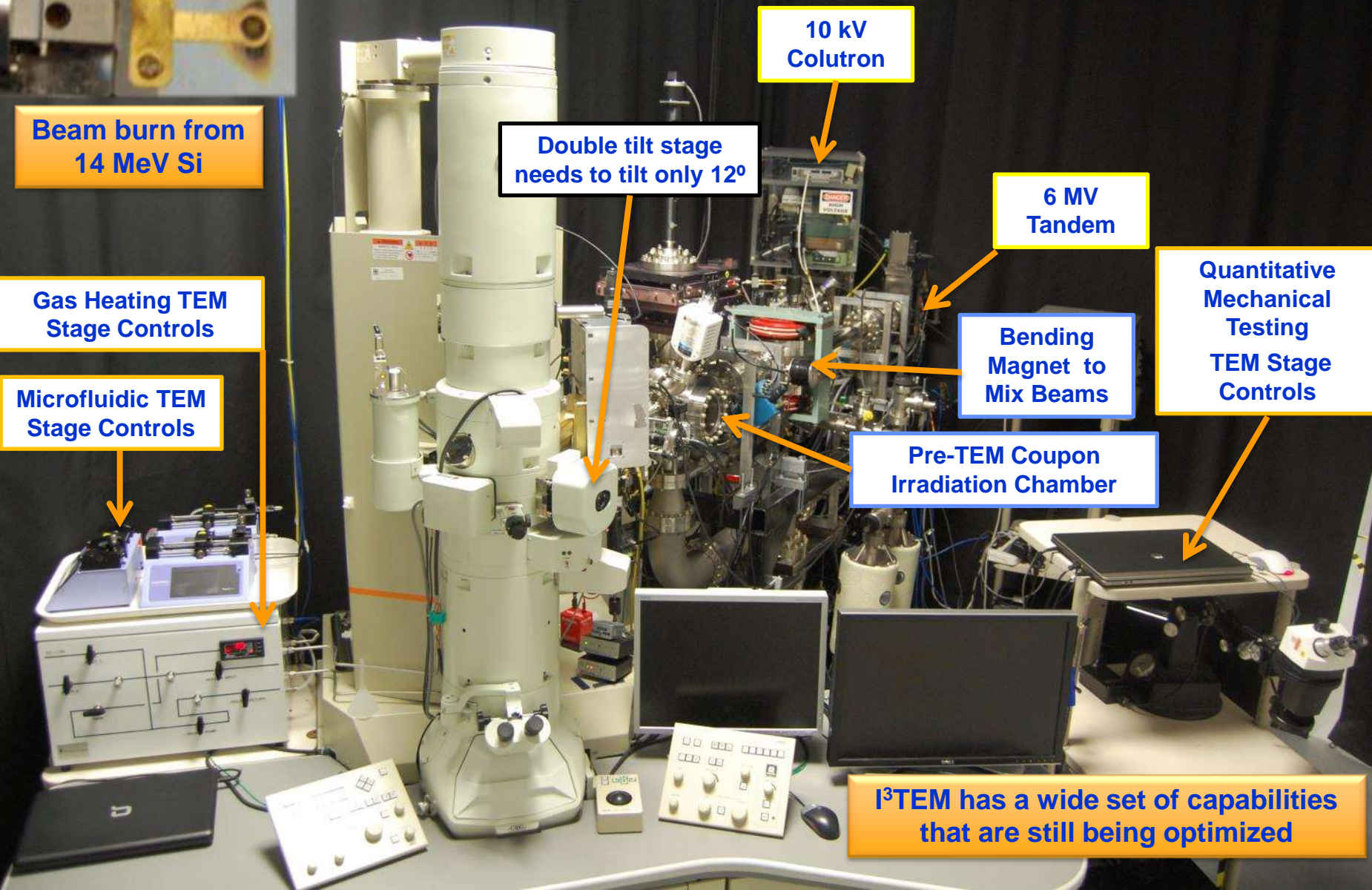
6 MV
Tandem

Bending
Magnet to
Mix Beams

Quantitative
Mechanical
Testing
TEM Stage
Controls

Pre-TEM Coupon
Irradiation Chamber

¹³TEM has a wide set of capabilities
that are still being optimized



Ion Species Attempted to Date

Collaborators: M. Steckbeck

1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	
<div>Hydrogen</div> <div>10/7 1 2011</div> <div>¹⁺ H 2.5</div> <div>1.008</div>																	<div>Helium</div> <div>4/18 2 2011</div> <div>¹⁺ He 2.5</div> <div>4.0026</div>	
<div>Lithium</div> <div>3</div> <div>Li</div> <div>6.94</div>	<div>Beryllium</div> <div>4</div> <div>Be</div> <div>9.0122</div>											<div>Boron</div> <div>5</div> <div>B</div> <div>10.81</div>	<div>Carbon</div> <div>7/08 6 2013</div> <div>¹⁺ C 1</div> <div>12.011</div>	<div>Nitrogen</div> <div>7</div> <div>N</div> <div>14.007</div>	<div>Oxygen</div> <div>8</div> <div>O</div> <div>15.999</div>	<div>Fluorine</div> <div>9</div> <div>F</div> <div>18.998</div>	<div>Neon</div> <div>10</div> <div>Ne</div> <div>20.180</div>	
<div>Sodium</div> <div>11</div> <div>Na</div> <div>22.990</div>	<div>Magnesium</div> <div>12</div> <div>Mg</div> <div>24.305</div>										<div>Aluminum</div> <div>7/18 13 2013</div> <div>³⁺ Al 3</div> <div>26.982</div>	<div>Silicon</div> <div>4/14 14 2011</div> <div>²⁺ Si 3</div> <div>28.085 5.5</div>	<div>Phosphorus</div> <div>15</div> <div>P</div> <div>30.974</div>	<div>Sulfur</div> <div>16</div> <div>S</div> <div>32.06</div>	<div>Chlorine</div> <div>17</div> <div>Cl</div> <div>35.45</div>	<div>Argon</div> <div>18</div> <div>Ar</div> <div>39.948</div>		
<div>Potassium</div> <div>19</div> <div>K</div> <div>39.098</div>	<div>Calcium</div> <div>20</div> <div>Ca</div> <div>40.078(4)</div>		<div>Scandium</div> <div>21</div> <div>Sc</div> <div>44.956</div>	<div>Titanium</div> <div>22</div> <div>Ti</div> <div>47.867</div>	<div>Vanadium</div> <div>23</div> <div>V</div> <div>50.942</div>	<div>Chromium</div> <div>24</div> <div>Cr</div> <div>51.996</div>	<div>Manganese</div> <div>25</div> <div>Mn</div> <div>54.938</div>	<div>Iron</div> <div>11/26 26 2012</div> <div>²⁺ Fe 3</div> <div>55.845(2)</div>	<div>Cobalt</div> <div>27</div> <div>Co</div> <div>58.933</div>	<div>Nickel</div> <div>11/4 28 2011</div> <div>³⁺ Ni 3.3</div> <div>58.693</div>	<div>Copper</div> <div>9/15 29 2011</div> <div>³⁺ Cu 3.3</div> <div>63.546(3)</div>	<div>Zinc</div> <div>30</div> <div>Zn</div> <div>65.38(2)</div>	<div>Gallium</div> <div>31</div> <div>Ga</div> <div>69.723</div>	<div>Germanium</div> <div>32</div> <div>Ge</div> <div>72.63</div>	<div>Arsenic</div> <div>33</div> <div>As</div> <div>74.922</div>	<div>Selenium</div> <div>34</div> <div>Se</div> <div>78.96(3)</div>	<div>Bromine</div> <div>35</div> <div>Br</div> <div>79.904</div>	<div>Krypton</div> <div>36</div> <div>Kr</div> <div>83.798(2)</div>
<div>Rubidium</div> <div>37</div> <div>Rb</div> <div>85.468</div>	<div>Sr</div> <div>38</div> <div>Sr</div> <div>87.62</div>		<div>Yttrium</div> <div>39</div> <div>Y</div> <div>88.906</div>	<div>Zirconium</div> <div>40</div> <div>Zr</div> <div>91.224(2)</div>	<div>Niobium</div> <div>41</div> <div>Nb</div> <div>92.906(2)</div>	<div>Molybdenum</div> <div>42</div> <div>Mo</div> <div>95.96(2)</div>	<div>Technetium</div> <div>43</div> <div>Tc</div> <div>[97.91]</div>	<div>Ruthenium</div> <div>44</div> <div>Ru</div> <div>101.07(2)</div>	<div>Rhodium</div> <div>45</div> <div>Rh</div> <div>102.91</div>	<div>Palladium</div> <div>46</div> <div>Pd</div> <div>106.42</div>	<div>Silver</div> <div>7/02 47 2013</div> <div>³⁺ Ag 2.6</div> <div>107.87</div>	<div>Cadmium</div> <div>48</div> <div>Cd</div> <div>112.41</div>	<div>Indium</div> <div>49</div> <div>In</div> <div>114.82</div>	<div>Tin</div> <div>50</div> <div>Sn</div> <div>118.71</div>	<div>Antimony</div> <div>51</div> <div>Sb</div> <div>121.76</div>	<div>Tellurium</div> <div>52</div> <div>Te</div> <div>127.60(3)</div>	<div>Iodine</div> <div>53</div> <div>I</div> <div>126.90</div>	<div>Xenon</div> <div>54</div> <div>Xe</div> <div>131.29</div>
<div>Caesium</div> <div>55</div> <div>Cs</div> <div>132.91</div>	<div>Barium</div> <div>56</div> <div>Ba</div> <div>137.33</div>	<div>57-70</div> <div>*</div>	<div>Lutetium</div> <div>71</div> <div>Lu</div> <div>174.97</div>	<div>Hafnium</div> <div>72</div> <div>Hf</div> <div>178.49(2)</div>	<div>Tantalum</div> <div>73</div> <div>Ta</div> <div>180.95</div>	<div>Tungsten</div> <div>6/25 74 2012</div> <div>⁴⁺ W 2</div> <div>183.84</div>	<div>Rhenium</div> <div>75</div> <div>Re</div> <div>186.21</div>	<div>Osmium</div> <div>76</div> <div>Os</div> <div>190.23(2)</div>	<div>Iridium</div> <div>77</div> <div>Ir</div> <div>192.22</div>	<div>Platinum</div> <div>78</div> <div>Pt</div> <div>195.08</div>	<div>Gold</div> <div>7/15 79 2011</div> <div>³⁺ Au 1.7</div> <div>196.97 3.35 4.5 6.5</div>	<div>Mercury</div> <div>80</div> <div>Hg</div> <div>200.59</div>	<div>Thallium</div> <div>81</div> <div>Tl</div> <div>204.38</div>	<div>Lead</div> <div>82</div> <div>Pb</div> <div>207.2</div>	<div>Bismuth</div> <div>83</div> <div>Bi</div> <div>208.98</div>	<div>Polonium</div> <div>84</div> <div>Po</div> <div>[209.98]</div>	<div>Astatine</div> <div>85</div> <div>At</div> <div>[209.99]</div>	<div>Radon</div> <div>86</div> <div>Rn</div> <div>[222.02]</div>
<div>Francium</div> <div>87</div> <div>Fr</div> <div>[223.02]</div>	<div>Radium</div> <div>88</div> <div>Ra</div> <div>[226.03]</div>	<div>89-102</div> <div>**</div>	<div>Lanthanum</div> <div>103</div> <div>La</div> <div>[262.11]</div>	<div>Rutherfordium</div> <div>104</div> <div>Rf</div> <div>[265.12]</div>	<div>Dubnium</div> <div>105</div> <div>Db</div> <div>[268.13]</div>	<div>Seaborgium</div> <div>106</div> <div>Sg</div> <div>[271.13]</div>	<div>Bohrium</div> <div>107</div> <div>Bh</div> <div>[270]</div>	<div>Hassium</div> <div>108</div> <div>Hs</div> <div>[277.15]</div>	<div>Meitnerium</div> <div>109</div> <div>Mt</div> <div>[276.15]</div>	<div>Darmstadtium</div> <div>110</div> <div>Ds</div> <div>[281.16]</div>	<div>Roentgenium</div> <div>111</div> <div>Rg</div> <div>[280.16]</div>	<div>Copernicium</div> <div>112</div> <div>Cn</div> <div>[285.17]</div>	<div>Ununtrium</div> <div>113</div> <div>Uut</div> <div>[284.18]</div>	<div>Flerovium</div> <div>114</div> <div>Fl</div> <div>[289.19]</div>	<div>Ununpentium</div> <div>115</div> <div>Uup</div> <div>[288.19]</div>	<div>Livermorium</div> <div>116</div> <div>Lv</div> <div>[293]</div>	<div>Ununseptium</div> <div>117</div> <div>Uus</div> <div>[294]</div>	<div>Ununoctium</div> <div>118</div> <div>Uuo</div> <div>[294]</div>

Successful Run

Unsuccessful Run

Key:

Month/day

10/07

Element Name

Atomic number

2011

Symbol

2.5

Atomic weight (mean relative mass)

Year of 1st Run

Current:

HIGH (>100 nA)

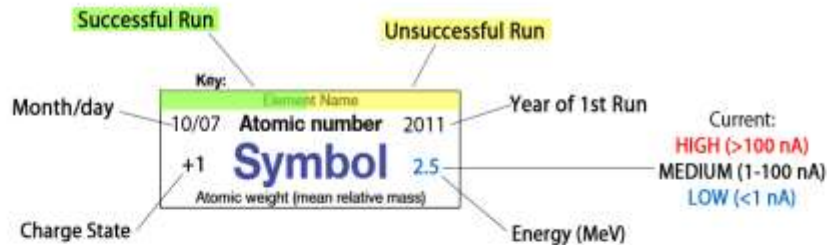
MEDIUM (1-100 nA)

LOW (<1 nA)

Charge State

+1

Energy (MeV)



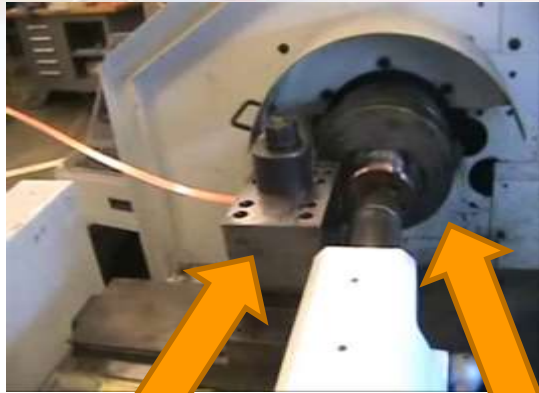
*lanthanoids

**actinoids

Lanthanum 57 ⁰ La 138.91	Cerium 58 ⁰ Ce 140.12	Praseodymium 59 ⁰ Pr 140.91	Neodymium 60 ⁰ Nd 144.24	Promethium 61 ⁰ Pm [144.91]	Samarium 62 ⁰ Sm 150.36(2)	Europium 63 ⁰ Eu 151.96	Gadolinium 64 ⁰ Gd 157.25(3)	Terbium 65 ⁰ Tb 158.93	Dysprosium 66 ⁰ Dy 162.50	Holmium 67 ⁰ Ho 164.93	Erbium 68 ⁰ Er 167.26	Thulium 69 ⁰ Tm 168.93	Ytterbium 70 ⁰ Yb 173.05
Actinium 89 ⁰ Ac [227.03]	Thorium 90 ⁰ Th 232.04	Protactinium 91 ⁰ Pa 231.04	Uranium 92 ⁰ U 238.03	Neptunium 93 ⁰ Np [237.05]	Plutonium 94 ⁰ Pu [244.06]	Americium 95 ⁰ Am [243.06]	Curium 96 ⁰ Cm [247.07]	Berkelium 97 ⁰ Bk [247.07]	Californium 98 ⁰ Cf [251.08]	Einsteinium 99 ⁰ Es [252.08]	Fermium 100 ⁰ Fm [257.10]	Mendelevium 101 ⁰ Md [268.10]	Nobelium 102 ⁰ No [259.10]

What Insight into Structural Stability is Gained from I³TEM Experiments?

Collaborators: O. El-Atwani, J. P. Allain, D. Buller, & J.A. Scott

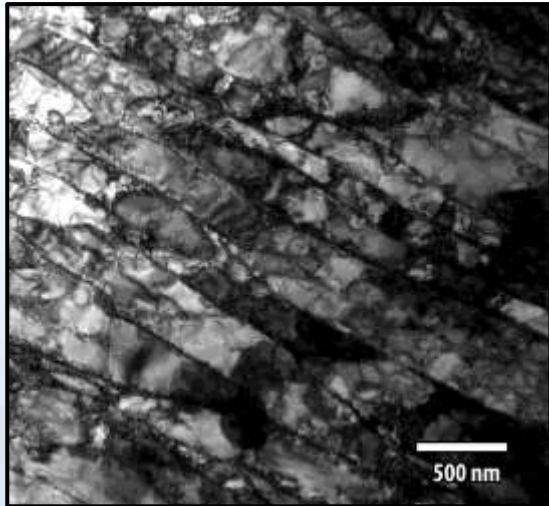


**Extrusion
machining tooling**

**Commercially
available lathe**

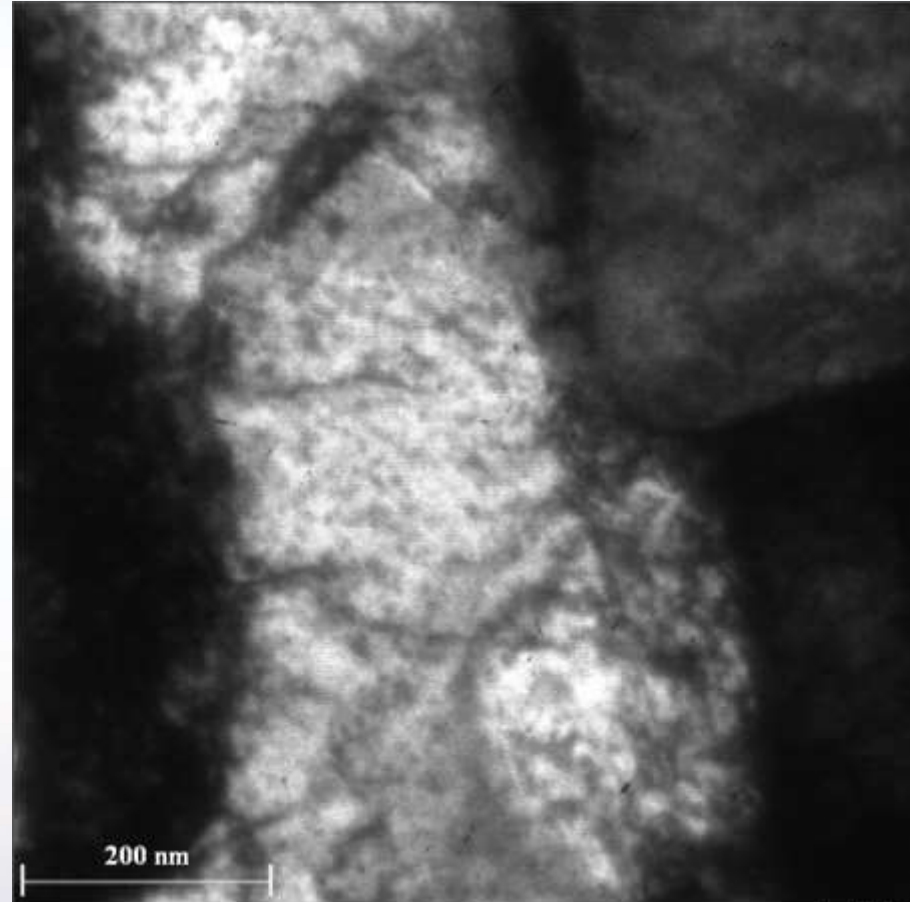
From space application
through nuclear reactor
cladding to waste
storage:

**Understanding
Radiation Damage is
Essential**



UFG Tungsten

- I³TEM W irradiation and He implantation of SPD-W developed for ITER applications



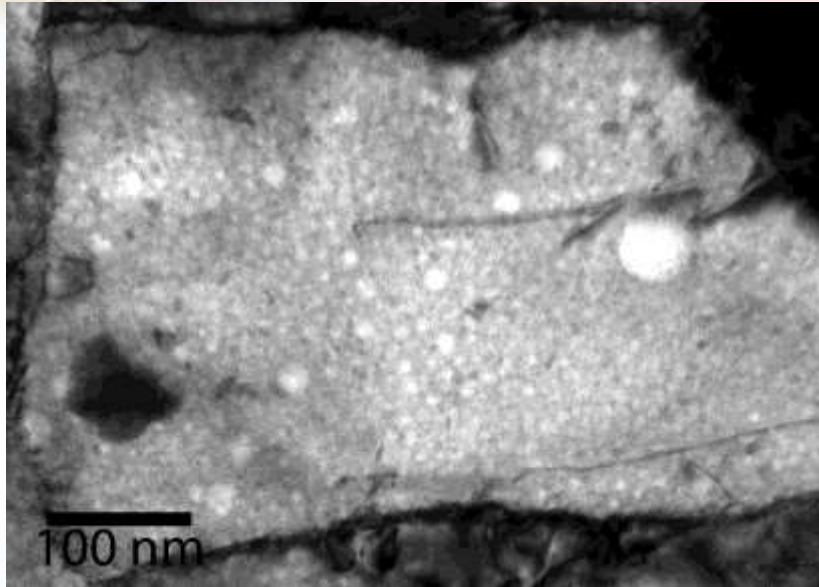
I³TEM is providing insight into:

- 1) Loop formation
- 2) Loop stability & migration
- 3) Rad & structural defect interactions

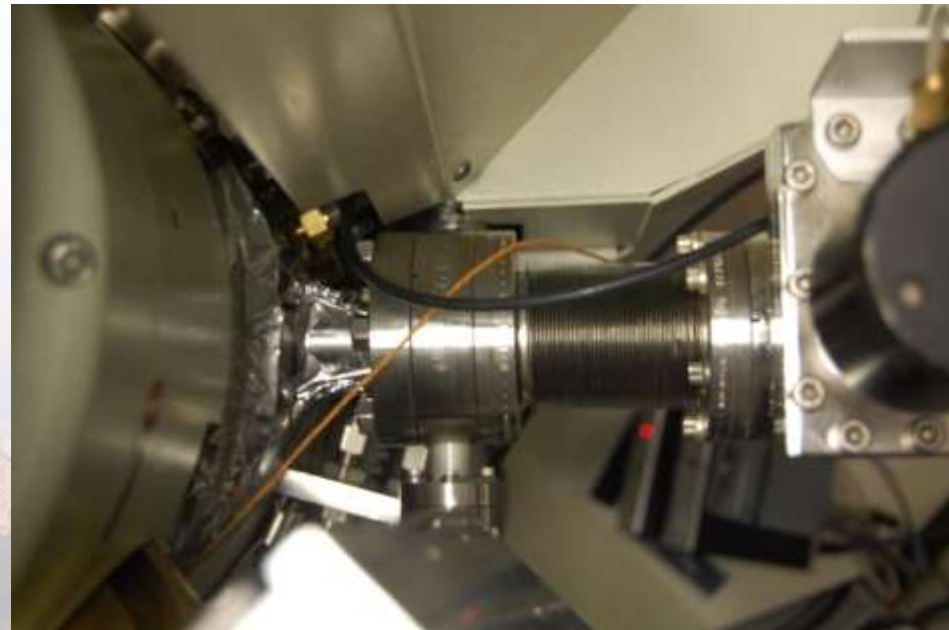
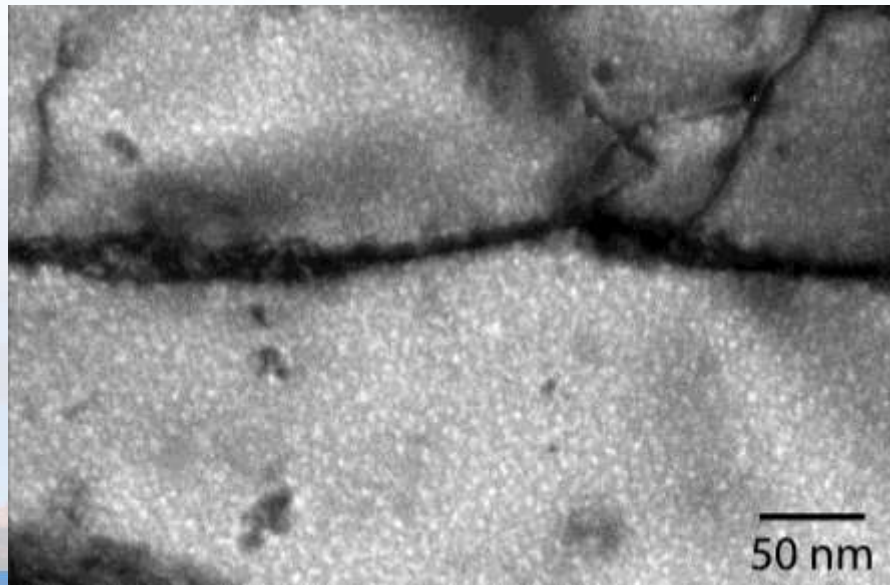


What Insight into Helium Bubble Formation is Gained from I³TEM Experiments?

Collaborators: O. El-Atwani, J. P. Allain, D. Buller, & J.A. Scott



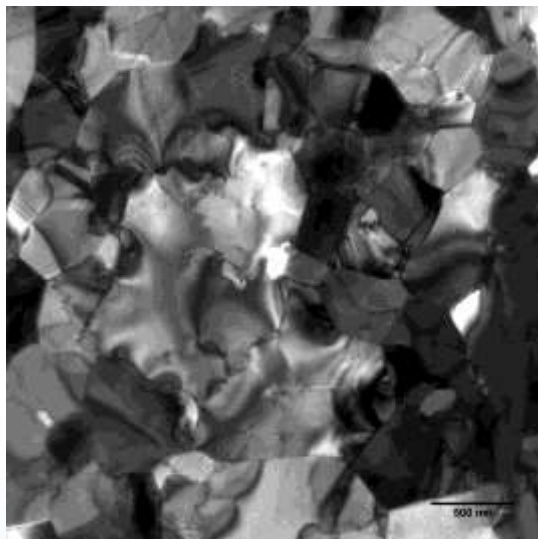
Electrostatic steering magnets were added into the bellows vicinal to the TEM. Should permit real time high magnification imaging of He bubbles.



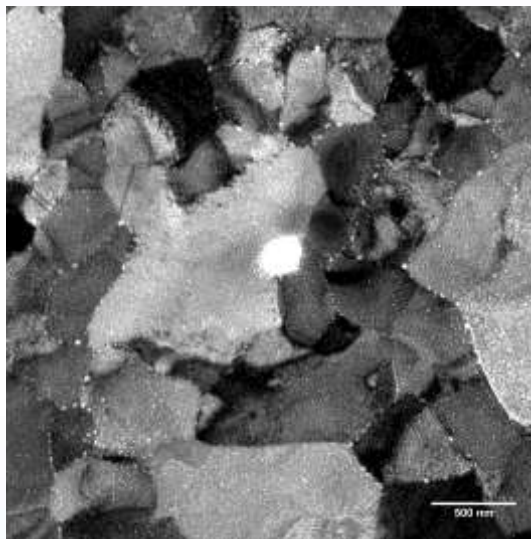
He Implantation and Subsequent Self-ion Irradiation of Au films

Collaborators: C. Chisholm & A.Minor

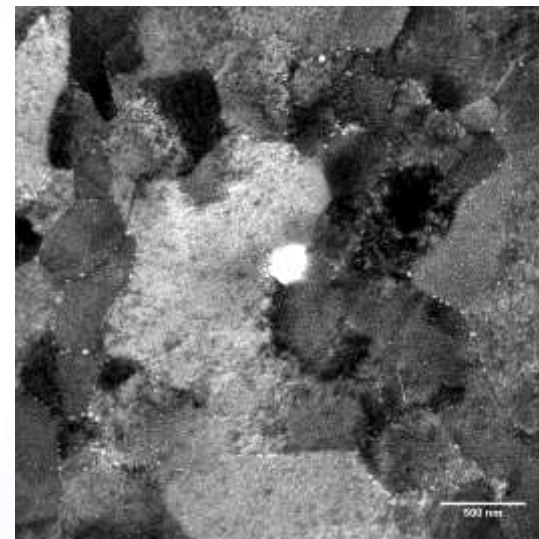
Unirradiated



1.5×10^{17} ions/cm² 10keV He²⁺

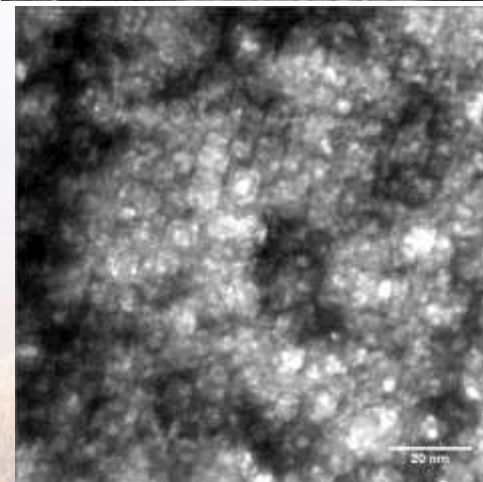
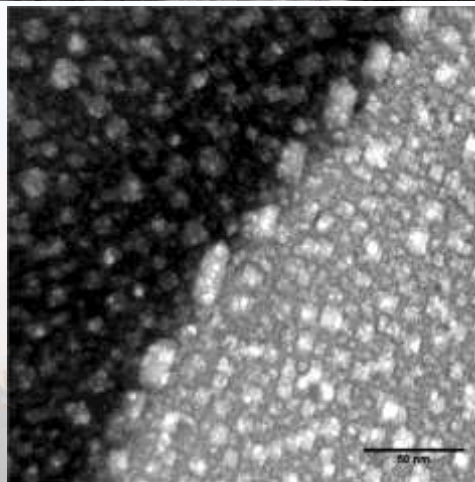


1.5×10^{17} ions/cm² 10keV He²⁺ then
 5.8×10^{10} ions/cm² 2.8MeV Au⁴⁺



Gold thin-film was
implanted with 10keV He²⁺
with a resulting porous
microstructure

The thin-film was then
irradiated with 2.8MeV Au⁴⁺,
adding dislocation loops



Sandia National Laboratories

In situ Observations of Single Cascade Events?

Collaborators: C. Chisholm, A. Kinghorn, & A. Minor

2.8MeV Au⁴⁺

Very preliminary observations suggest that single ion strikes can be observed during self-ion irradiation of Au thin foils.

Current work is underway to confirm that each event is a single ion strike.

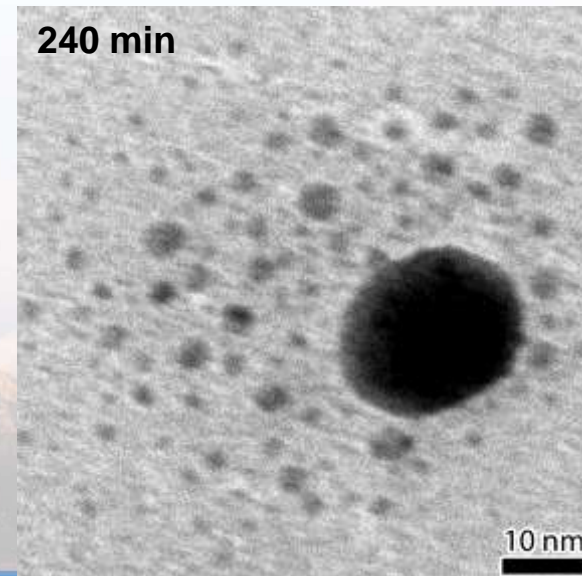
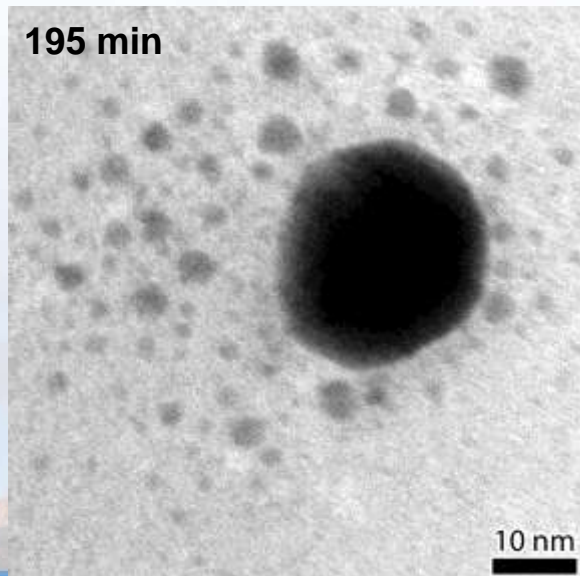
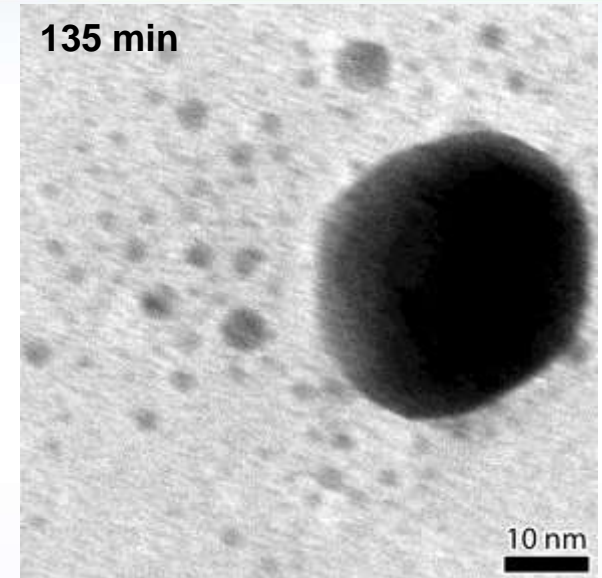
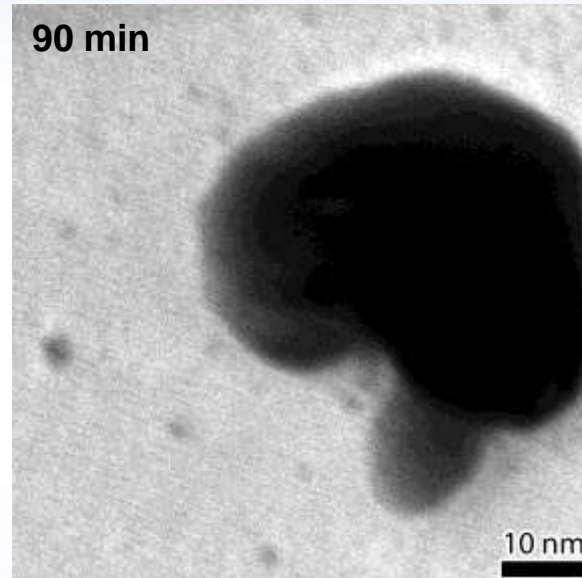
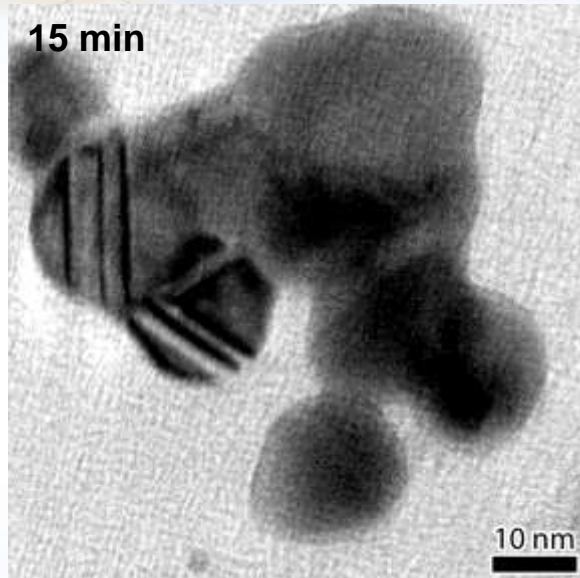
100 nm



Sandia National Laboratories

Au NP Irradiated with 100 nA of 3 MeV Cu³⁺

Collaborators: S.H. Pratt



Over 4 hours
a group of Au NP
coalesced and smaller
particles sputtered off,
then grew



Sandia National Laboratories

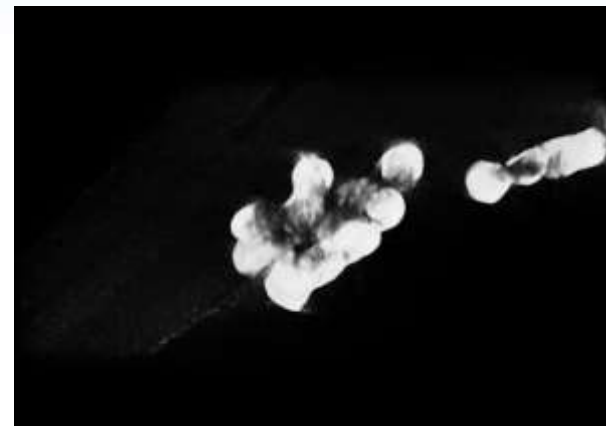
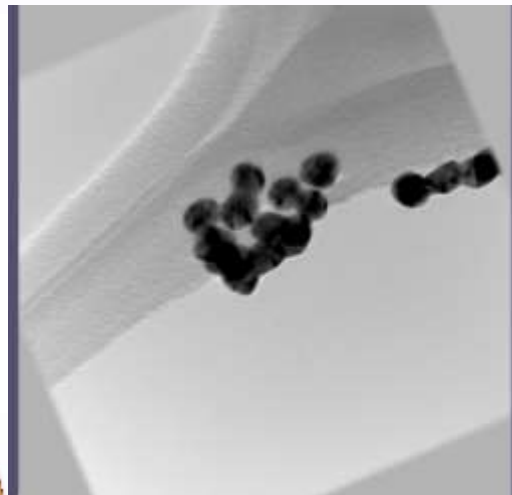
Advanced Microscopy Techniques Applied to Nanoparticles in Radiation Environments

Collaborators: S.H. Pratt

In situ Ion Irradiation TEM (I³TEM)

Aligned Au NP tilt series -
unirradiated

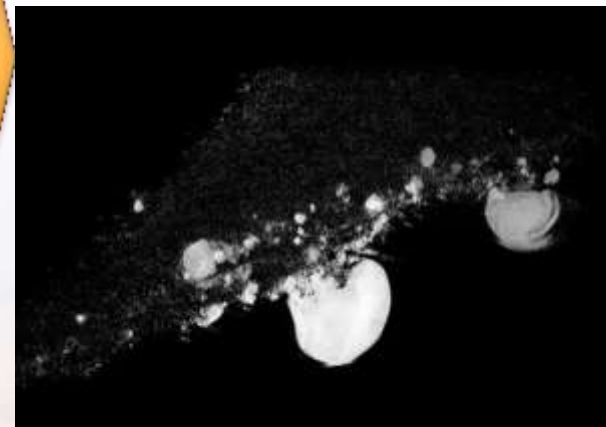
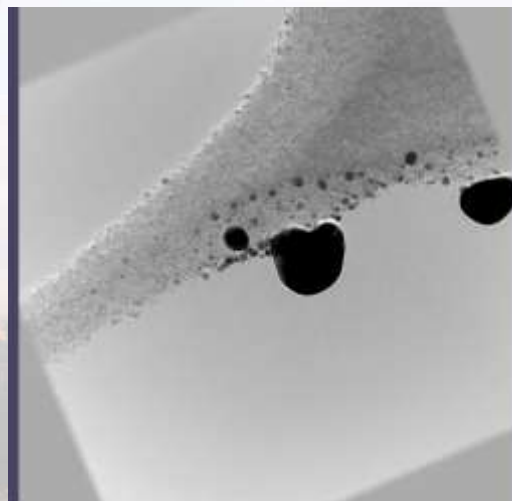
Unirradiated Au NP model



Hummingbird
tomography stage

Aligned Au NP tilt series -
irradiated

Irradiated Au NP model



The application of advanced
microscopy techniques to
extreme environments provides
exciting new research directions



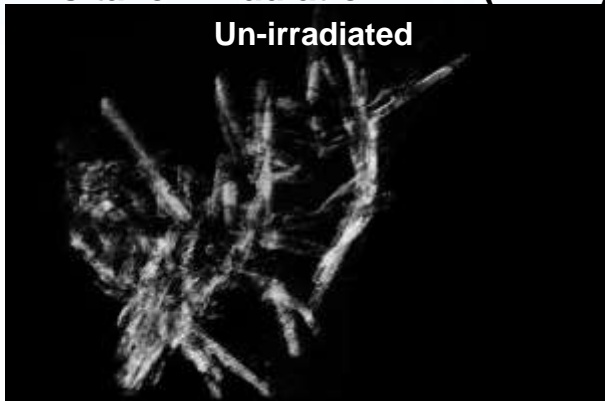
Radiation Tolerance is Needed in Advanced Scintillators for Non-proliferation Applications

Contributors: S.H. Pratt, B.A. Hernandez-Sanchez, T. Boyle

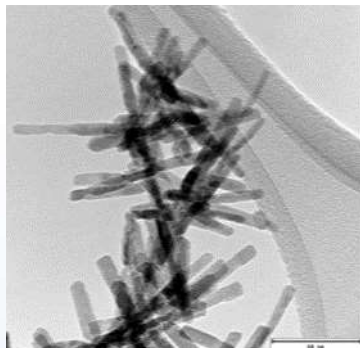


In situ Ion Irradiation TEM (I³TEM)

Un-irradiated

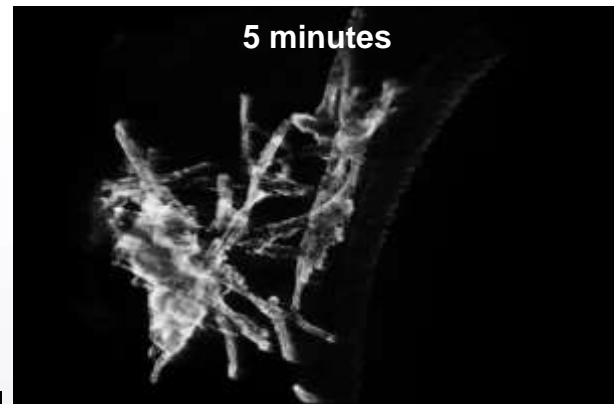


Tomography of Irradiated CdWO₄:
3 MeV Cu³⁺ at ~30 nA

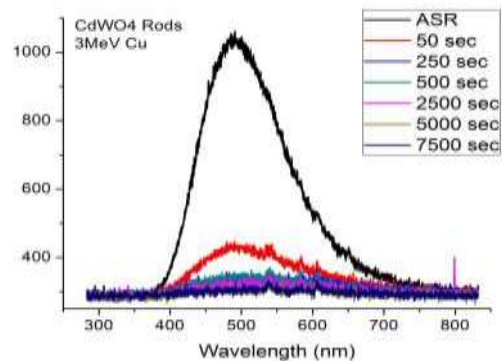
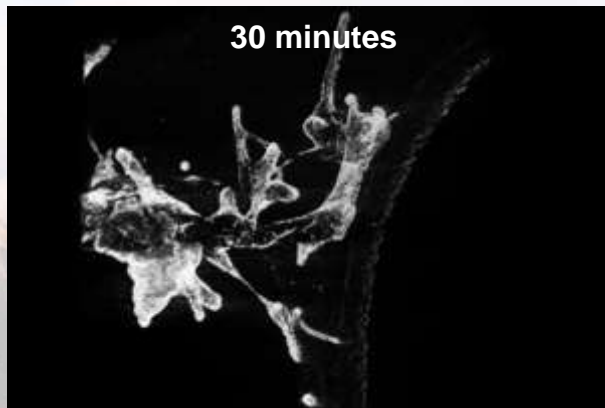


High-Z nanoparticles
(CdWO₄) are promising, but
are radiation sensitive

5 minutes



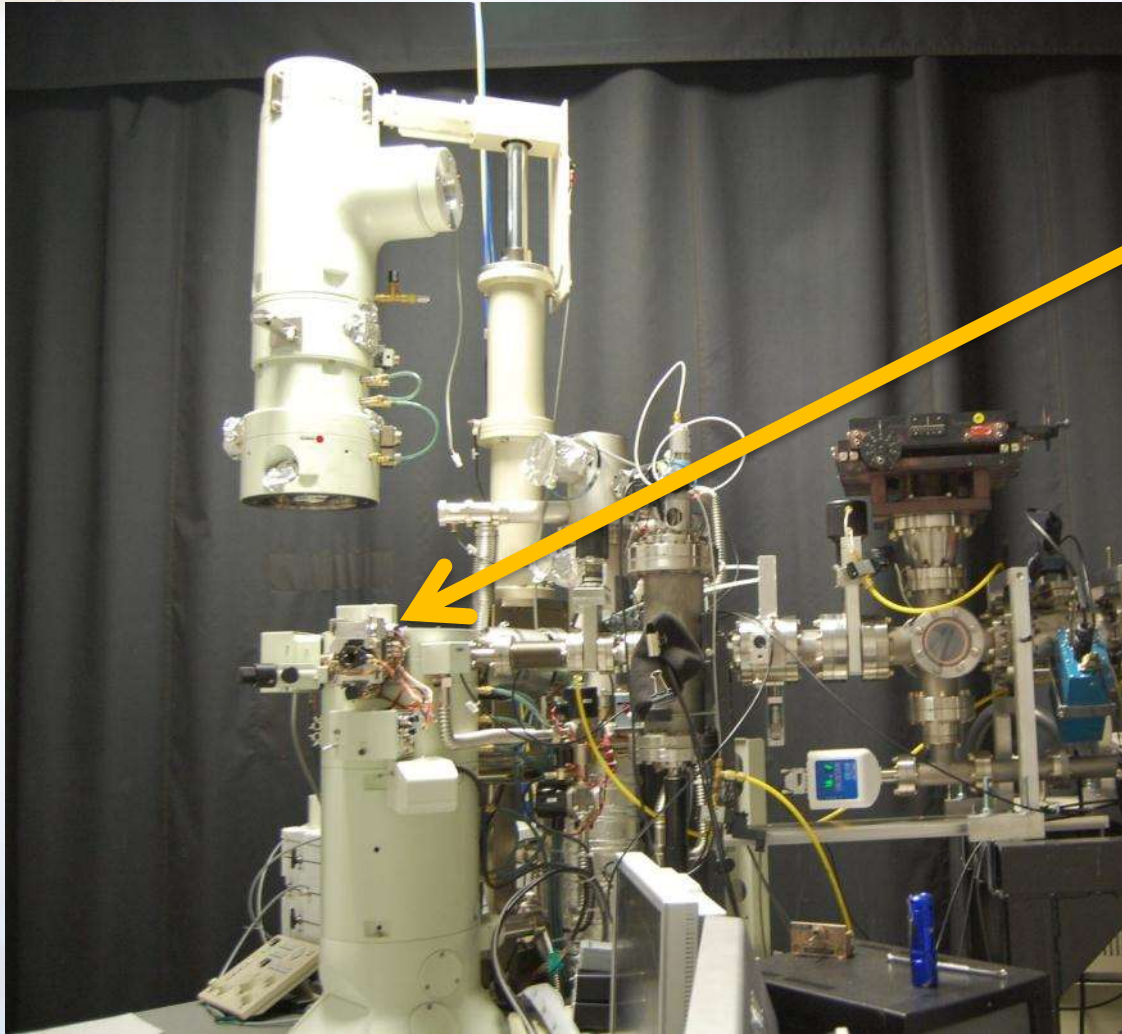
30 minutes



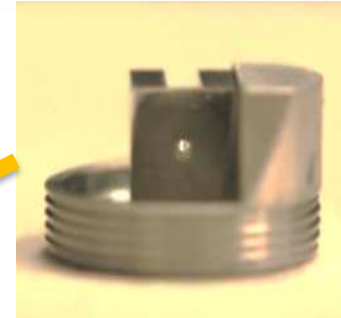
Sandia National Laboratories

In situ TEM Luminescence

Collaborators: D. Masiel



Optical Mirror in TEM



First IBIL in TEM



Optical Pathway in an I³TEM

- Angled mirror with bore hole for the electron path was installed.
- Mirror is located on top of the objective polepiece
- Port was constructed with thick leaded glass to permit light through, but limited x-rays created by ion or electron beams.
- Permits *in situ* IBIL.

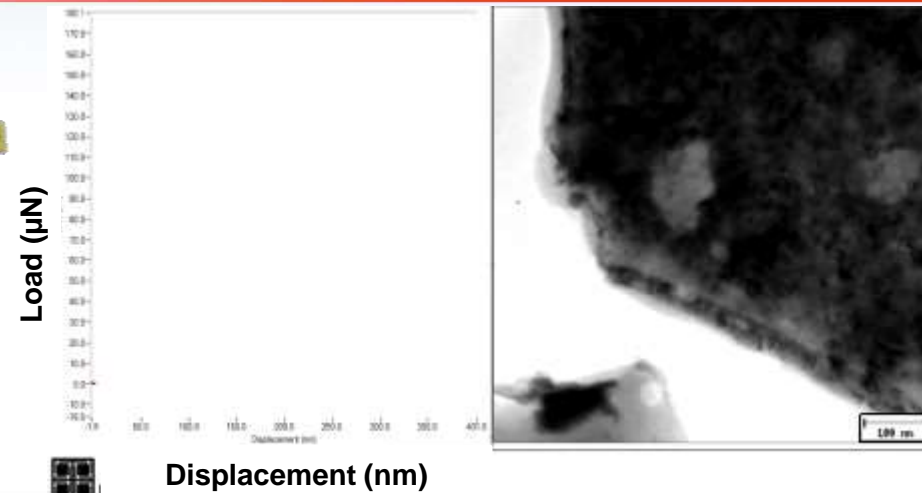
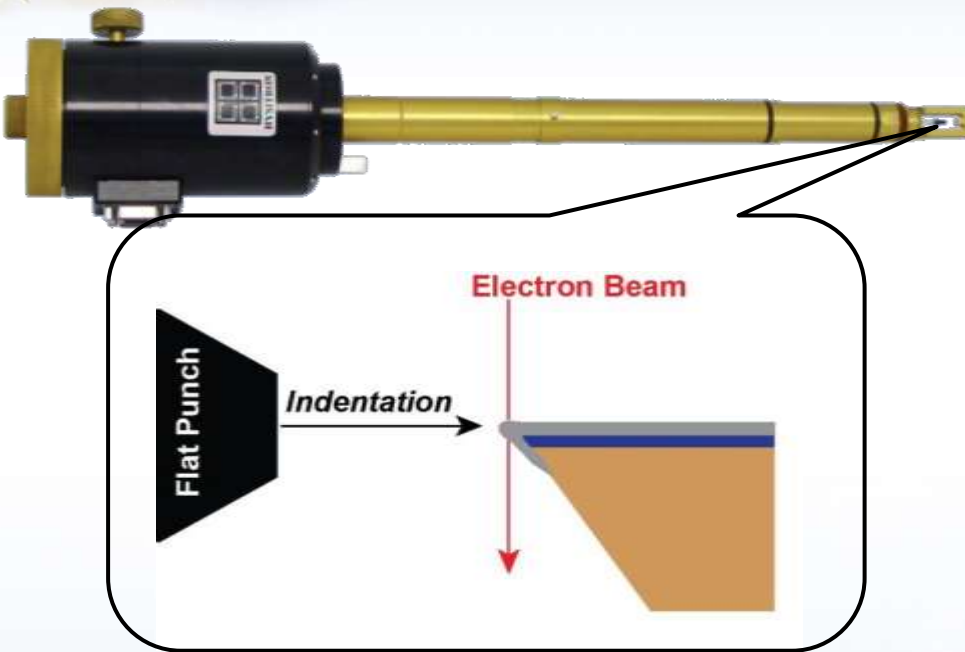
An optical port was added to the I³TEM, which permits *in situ* TEM luminescence studies



Sandia National Laboratories

In situ TEM Quantitative Mechanical Testing

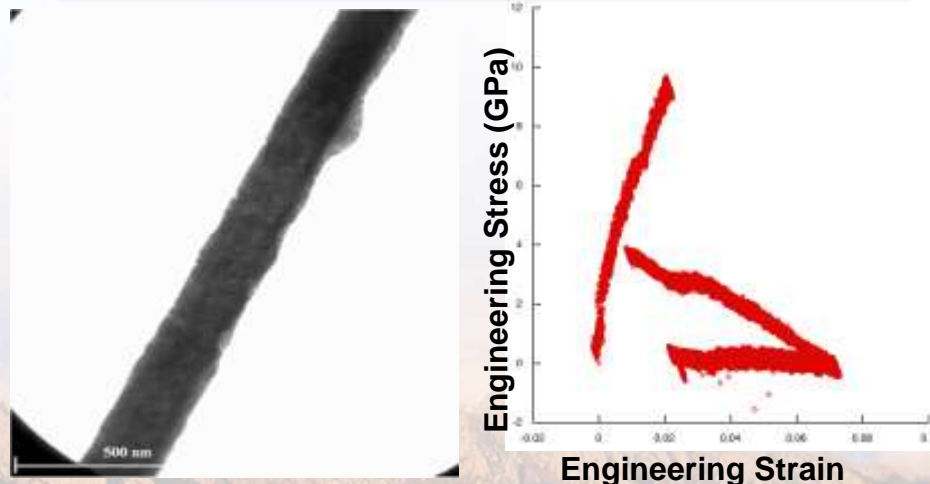
Contributors: J. Sharon, B. L. Boyce, C. Chisholm, H. Bei, E.P. George, P. Hosemann, A.M. Minor, & Hysitron Inc.



Fundamentals of Mechanical Properties

Range of Mechanical Testing Techniques

- Indentation
- Compression
- Tension
- Bending
- Wear
- Fatigue
- Creep



We have started looking at the effects of ion irradiation on mechanical properties



In situ TEM Gas Environments

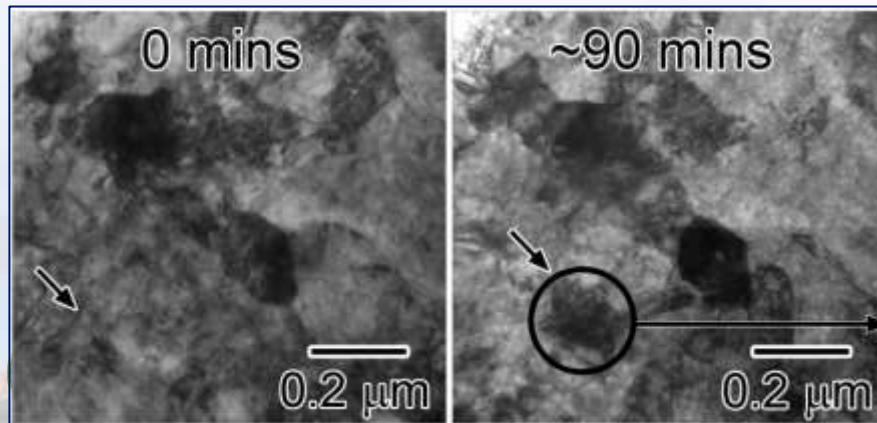
Contributors: B.G. Clark, S. Rajasekhara, P.J. Cappillino, B.W. Jacobs, M.A. Hekmaty, D.B. Robinson, L.R. Parent, I. Arslan. & Protochips, Inc.

Vapor-Phase Heating TEM Stage

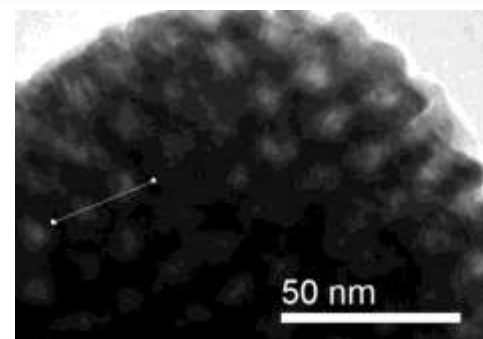
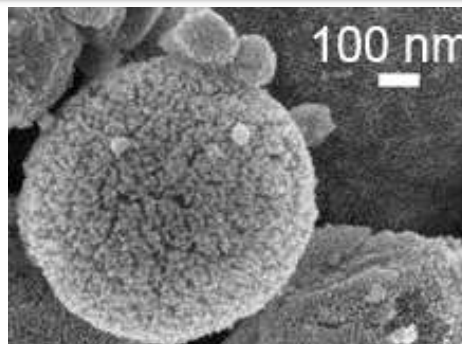
- Compatible with a range of gases
- *In situ* resistive heating
- Continuous observation of the reaction channel
- Chamber dimensions are controllable
- Compatible with MS and other analytical tools



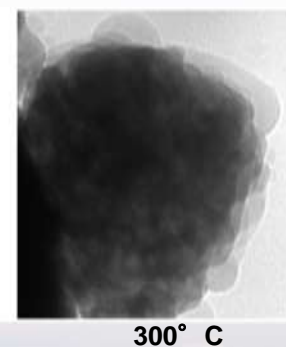
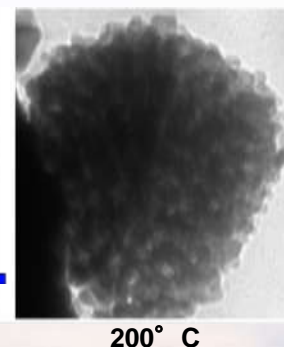
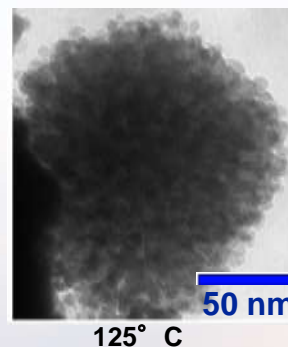
Absolute hydrogen pressure: 327 torr,
Ramp rate: 1 °C/s, Final temperature: ~ 400 C,



New *in situ* atmospheric heating experiments provide great insight into range of environmental studies



- 1 atm H₂ after several pulses to specified temp.



Even greater potential is envisioned when it is combined with *in situ* ion irradiation.



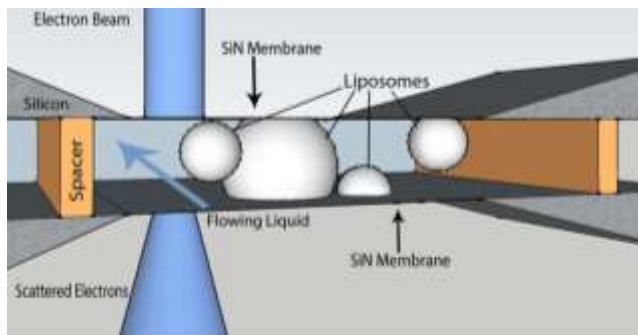
Sandia National Laboratories

In situ TEM microfluidic Enviroments

Contributors: S.H. Pratt, E. Carnes, J. Brinker, D. Sasaki, D. Gross, J. Kacher, I.M. Robertson & Protochips Inc.

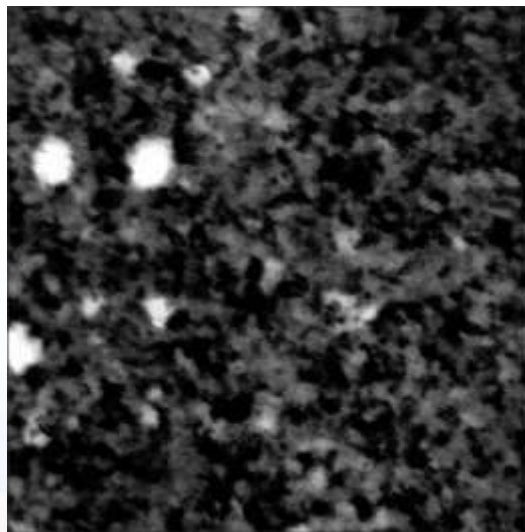
Microfluidic Stage

- Mixing of two or more channels
- Continuous observation of the reaction channel
- Chamber dimensions are controllable



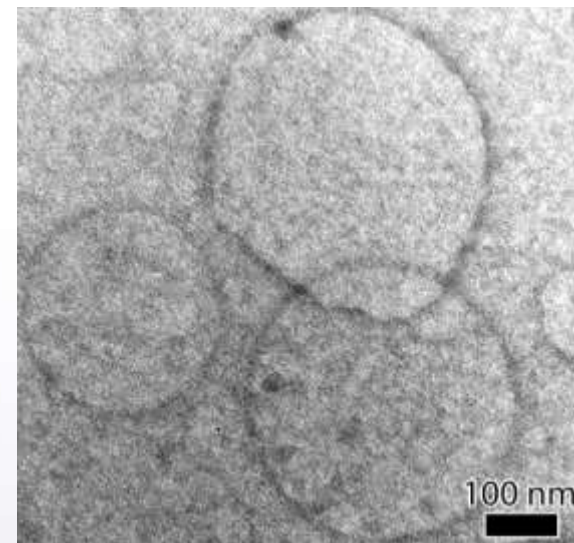
In situ microfluidic TEM can provide insight in events as diverse as corrosion and drug delivery

Fe Corrosion



Dilute flow of acetic acid over
99.95% nc-PLD Fe

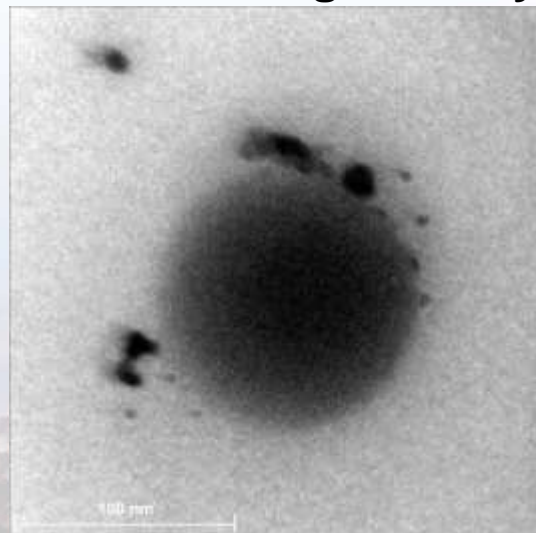
Liposomes in Water



Liposomes imaged in flowing
aqueous channel

Protocell Drug Delivery

Liposome
encapsulated
Silica
destroyed by
the electron
beam in
aqueous
environment



We hope to go a step further and combine it with
ion implantation and irradiation environments



Sandia National Laboratories

Summary

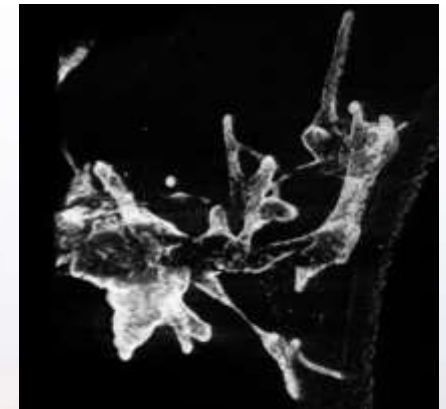
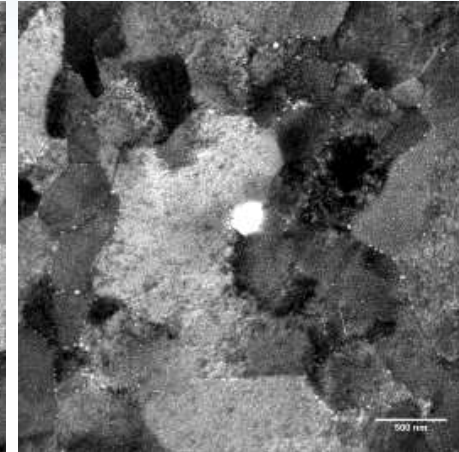
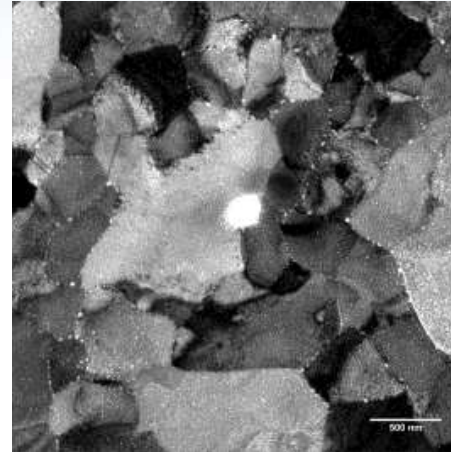
Current Status of the I³TEM

- Perform real time heavy ion irradiation
- Ion implantation capability is operational
 - Still being optimized
- Can operate at elevated temperatures
- Combined with tomography
 - Provides 4D insight as a function of dose

Future Combinations with I³TEM

- Quantitative mechanical property testing
- High temperature gas environments
- Microfluidic environments
- ...

Sandia's I³TEM, although still under development, is testing the frontiers of *in situ* ion irradiation microscopy and providing a wealth of interesting initial observations along the way



Contributors & Collaborators:

J. Hinks, S.H. Pratt, A. Kinghorn, M. Steckbeck, B.A. Hernandez-Sanchez, T. Boyle, E. Carnes, J. Brinker, D. Sasaki, D. Gross, J. Kacher, I.M. Robertson, Protochips, B.G. Clark, S. Rajasekhara, P.J. Cappillino, B.W. Jacobs, M.A. Hekmaty, D.B. Robinson, L.R. Parent, I. Arslan, J. Sharon, B. L. Boyce, C. Chisholm, H. Bei, E.P. George, P. Hosemann, A.M. Minor, Hysitron, & D. Masiel



Sandia National Laboratories is a multi-program laboratory managed and operated by Sandia Corporation, a wholly owned subsidiary of Lockheed Martin Corporation, for the U.S. Department of Energy's National Nuclear Security Administration under contract DE-AC04-94AL85000.



Sandia National Laboratories