Centre of Excellence for Nuclear Materials

Workshop

Materials Innovation for Nuclear Optimized Systems

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Lucile BECK
CEA (France)

CEA Charged Particle Irradiation Facilities for Nuclear Material studies

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CEA Charged Particle Irradiation Facilities for Nuclear Material Studies

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To study the effects of radiation on nuclear materials, the CEA uses various irradiation facilities producing neutrons or charges particles. Neutron facilities have the great advantage to directly produce neutron damages, but have also some drawbacks. The samples are activated and consequently their characterizations need dedicated hot cells, the reactors have fixed experimental parameters and the experiments are long and costly.

In order to better understand the mechanisms of radiation damage, experimental simulation can be also conducted with charged particles. In this case, experimental irradiation conditions (temperature, dose, flux, energy) are well controlled and the irradiated samples can be characterized with conventional analytical methods. This presentation will describe the CEA facilities devoted to ion or electron irradiations for material studies. Examples of irradiations performed at JANNUS (Joint Accelerators for Nano-science and NUClear Simulation) will be detailed (Fig. 1).

Fig. 1: Schematic view of the JANNUS platform at Saclay.

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CEA charged particle irradiation facilities for nuclear material studies

Lucile Beck

CEA/DMN/SRMP/Jannus Laboratory - Saclay
with the contributions of
B. Boizot (LSI), S. Bouffard & M. Toulemonde (CIMAP)
J. Henry & A. Jankowiak (SRMA)
P. Trocellier, S. Pellegrino, S. Miro, Y. Serruys (JANNUS)
X. Deschanels (ICSM)
PART 1 : CEA charged particle irradiation facilities

Ions

• CIMAP - Caen (CEA/DSM, CNRS/INP, ENSICAEN, Caen University)
• JANNUS – Saclay (CEA/DEN/DMN/SRMP)
  [GIS with JANNUS – Orsay (CNRS, Orsay University)]

Electrons

• LSI - Palaiseau (CEA/DSM, CNRS/INP, Ecole Polytechnique)
• THT – Saclay (CEA/DEN/DMN/SRMA)

• Located near Paris and in Caen
• Two CEA divisions: Materials Sciences Division (DSM)
  Nuclear Energy Division (DEN)
• in collaboration with CNRS and Universities
EMIR network
a national network of accelerators dedicated to material irradiation

EMIR network gathers the French facilities for material irradiation
- to promote the material research with the accelerators
- to meet for scientific challenge
  - the prediction of the ageing of nuclear materials
    - to achieve reliable simulations of the radiation damage through the study of model systems
  - to develop irradiation as a tool for
    - nano-structuration of materials
    - solid state physics
    - the physics of semiconductors
- to provide academic and industrial researchers access to irradiation facilities and online characterization
- to assess the need for development facilities
- to ensure the visibility of these facilities

A call for proposal of experiment every year: http://emir.in2p3.fr/EMIR-network
Next call: October 2013
Large choice of particles and energies for different interactions

Scheme of energy deposition of an ion in matter / electrons in matter

- Low energy ions (1-10 keV/u)
- Swift ions (1 to 100 MeV/u)
- MeV electrons

Elastic interactions with atom ballistic displacements

Inelastic interactions with electrons

© M. Toulemonde
Access to the CEA charged particle irradiation facilities

## Ions

<table>
<thead>
<tr>
<th>Facility</th>
<th>Beam Type</th>
<th>Energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>SRMP JANNUS-Saclay</td>
<td>H → Bi</td>
<td>0.5 – 30 MeV</td>
</tr>
<tr>
<td>Japet</td>
<td>H → Bi</td>
<td>12 MeV</td>
</tr>
<tr>
<td>Yvette</td>
<td>H, $^2$H, $^3$He, $^4$He</td>
<td>0.5 – 2.5 MeV</td>
</tr>
<tr>
<td>CSNSM JANNUS-Orsay</td>
<td>H → Bi</td>
<td>0.5 – 15 MeV</td>
</tr>
<tr>
<td>ARAMIS</td>
<td>H → Bi</td>
<td>5 – 670 keV</td>
</tr>
</tbody>
</table>

## Electrons

<table>
<thead>
<tr>
<th>Facility</th>
<th>Beam Type</th>
<th>Energy</th>
</tr>
</thead>
<tbody>
<tr>
<td>LSI</td>
<td>e-</td>
<td>0.15 – 2.5 MeV</td>
</tr>
<tr>
<td>SIRIUS</td>
<td>e-</td>
<td>1.2 MeV</td>
</tr>
<tr>
<td>SRMA</td>
<td>HVEM</td>
<td>e-</td>
</tr>
</tbody>
</table>

+ TEM
**Cimap-Caen:** medium to heavy ions – MeV energy range

Scientific themes:
- Physics of the ion – matter interaction
- Mechanisms of energy transfer,
- Damage mechanisms and structural modifications in materials,
- Simulation of the nuclear material behaviour under irradiation,
- Nanostructured by irradiation,
- Radiation chemistry
- Radiobiology

<table>
<thead>
<tr>
<th>Facility</th>
<th>Ion</th>
<th>Energy Range</th>
<th>Operating Conditions</th>
<th>Instruments</th>
</tr>
</thead>
<tbody>
<tr>
<td>GANIL</td>
<td>C, U</td>
<td>0.3 – 1 MeV/A</td>
<td>under vacuum 8 – 1100 K</td>
<td>X-ray diffractometer, Infrared spectrometer, Gas analysis</td>
</tr>
<tr>
<td>IRRSUD</td>
<td>C, U</td>
<td>4.5 – 13 MeV/A</td>
<td>under vacuum or gas 8 – 1400 K</td>
<td>X-ray diffractometer, Infrared spectrometer, Gas analysis</td>
</tr>
</tbody>
</table>
JANNUS-Saclay: 3 joint accelerators
Low to heavy ions – MeV energy range

<table>
<thead>
<tr>
<th></th>
<th>Beam Species</th>
<th>Energy Range</th>
<th>Operating Environment</th>
<th>Research Themes</th>
</tr>
</thead>
<tbody>
<tr>
<td>Epiméthée</td>
<td>H → Bi</td>
<td>0.5 – 30 MeV</td>
<td>under vacuum</td>
<td>NRA – RBS, ERDA – PIXE</td>
</tr>
<tr>
<td>.Iapet</td>
<td>H → Bi</td>
<td>tandem 2 MV</td>
<td>under vacuum 77 – 800 K</td>
<td></td>
</tr>
<tr>
<td>Yvette</td>
<td>H, ²H, ³He, ⁴He</td>
<td>0.5 – 2.5 MeV</td>
<td>under vacuum</td>
<td></td>
</tr>
</tbody>
</table>

Scientific theamatics
- Microstructure evolution under irradiation for nuclear materials
- Synergetic effects of damage and Helium and Hydrogen production
- Atomic transport phenomena under irradiation
- Controlled modification by irradiation of the material properties
- Teaching and formation with the accelerators
## LSI-Palaiseau: MeV energy electrons

### LSI - Palaiseau

<table>
<thead>
<tr>
<th>SIRIUS</th>
<th>e-</th>
<th>0.15 – 2.5 MeV</th>
<th>High doses (GGy)</th>
<th>Large area 20 – 600 K</th>
<th>Optical spectroscopies</th>
<th>Micro-electrochemistry</th>
<th>Electrical measurements</th>
<th>Gas emission</th>
</tr>
</thead>
</table>

### Scientific thematics

- **Irradiation effects on material properties**
- **Material ageing under irradiation**
- **Characterization of induced defects (defect type, displacement threshold)**
- **Radio-grafting of polymers**
- **Corrosion under irradiation**
SRMA-Saclay: MeV energy electrons

SRMA - Saclay

<table>
<thead>
<tr>
<th>HVEM</th>
<th>e-</th>
<th>0.3 – 1.2 MeV</th>
<th>under vacuum</th>
<th>TEM</th>
</tr>
</thead>
</table>

Scientific thematics
- Microstructure evolution under irradiation for nuclear materials
- Effects of the high damage rate
Example 1: Mesoporous silica gels for applications in the field of nuclear waste

X. Deschanels, S. Dourdain and coll.
ICSM, Marcoule

Separation chemistry
Waste management

Sample characteristics
- Si substrate
- Some samples filled with C and Cs to simulate a radionuclide
- Layer thickness: ~100nm

SBA15: Structure 2D hexagonale
Structure 3D hexagonale

dpore=5nm

dpore = 2nm
Example 1: Mesoporous SiO$_2$ gel

X. Deschanels, S. Dourdain and coll.  
ICSM, Marcoule

Inelastic processes  
*Ganil, CEMHTI*

Ballistic processes  
*Jannus*

**Irradiations conditions**

<table>
<thead>
<tr>
<th>Inelastic processes</th>
<th>Xe : 92 MeV</th>
<th>$1\times10^{13}$ cm$^{-2}$</th>
<th>$T_{\text{amb.}}$</th>
<th>$(dE/dx)_e=9$ keV/nm</th>
</tr>
</thead>
</table>

| 20 MeV Ar | $S_e=3.4$ keV/nm |
| 0.5 MeV He | $S_e=0.26$ keV/nm | @ RT |
Example 1: Mesoporous SiO$_2$ gel

- Parameters of the coating
  - Thickness decrease
  - Porosity volume increase

Destructuration of the silica network under Xe 92 MeV

@ GANIL
Example 1: Mesoporous SiO$_2$ gel

- $d_p = 5$ nm
- $d \approx 100$ nm

0.5 MeV He, 20 MeV Ar @ JANNUS

- Shrinkage of the film
- Slight organisation of the film

- Complete collapse
- Densification and loss in the organisation of the thin films even for $S_e$ lower than track threshold

He irradiation
- 0.5 MeV He, $10^{10}$ cm$^{-2}$

Ar irradiation
- 20 MeV Ar, $10^{13}$ cm$^{-2}$
Example 2: *In-situ* mechanical test

IRRSUD line GANIL Caen  
Épiméthée E3 line JANNUS Saclay

⇒ Damage effect for both electronic and nuclear slowing down regimes

A. Jankowiak, Ch. Colin (CEA/DM/\textregistered SRMA) et coll.
Example 2: *In-situ* mechanical test

**Development of a *in-situ* tensile test device**

- Tensile test on single SiC fiber up to 5N;
- Usable for other type of materials;
- Gauge length: 25 mm;
- *In-situ* measurements of the fiber diameter possible;
- Operated in vacuum: $10^{-6}$ to $10^{-7}$mbar;
- Tensile tests (creep tests) from 25°C à 1800°C;
- Graphite-grip method: cold grips and uniform temperature;
- Implementable on every standard type of beam line;
Example 2: *In-situ* mechanical test

*In situ* tensile tests on SiC fibers under ion beam

IRRSUD line GANIL Caen $\text{Xe}^{23+} \ 92\text{ MeV}$

Épiméthée E3 line JANNUS Saclay $\text{C}^+ \ 9$ and $12\text{ MeV}$

**Fiber at 1000 °C and 300MPa**

- Strain rate variation with flux intensity;
- Residual stain at 1000 °C < 0.1 % for $2.1 \times 10^{12} \ 92\text{ MeV} \ \text{Xe}^{23+} \text{ ions/cm}^2$ and $3.1 \times 10^{16} \ 12\text{ MeV} \ \text{C}^+ \text{ ions/cm}^2$

**Previous works carried out at RT** (irradiation-induced swelling study)

- Residual strain at RT ~ 0.50% % for $5.1 \times 10^{14} \ 92\text{ MeV} \ \text{Xe}^{23+} \text{ ions/cm}^2$ and ~0.45 % for $10^{17} \ 12\text{ MeV} \ \text{C}^+ \text{ ions/cm}^2$
- Annealing test at 1800°C: residual strain almost disappears completely
PhD thesis of S. Pellegrino (JANNUS): “Irradiation effects in carbides (SiC, TiC and ZrC) and synergetic effects of electronic and nuclear energy losses.”

GANIL-SME
Pb 900 MeV
High Energy
\((dE/dx)_{\text{nuc.}} < (dE/dx)_{\text{elec.}}\)

JANNUS
Au 1.2 MeV
Low Energy
\((dE/dx)_{\text{nuc.}} > (dE/dx)_{\text{elec.}}\)
DAMAGE CHARACTERIZATION WITH RBS in CHANNELING conditions (CSNSM-Orsay)

**SiC**
- Gold - 1.2 MeV
- Amorphization (single crystal in random orientation)
- Single crystal with damage
- Single crystal (oriented)

**TiC**
- Gold - 1.2 MeV

**ZrC**
- Gold - 1.2 MeV

Weak effect of electronic energy loss in SiC

Almost no effect in ZrC and TiC until $1.7 \times 10^{14}$ ions/cm² (strong difference with SiC)

**Au 1.2 MeV**

**Pb 900 MeV**

CEA – DEN

**SiC**

**TiC**

**ZrC**
EXAMPLE 4: ODS (OXIDE DISPERSED STRENGTHENED) STEELS

M. J. Fluss, S. Tumey, L. Hsiung (LLNL) and A. Kimura (Kyoto University)

Dual beam irradiation

ODS (K3 and MA957) Fe-Cr 14%

\[ {^{56}\text{Fe}}^{8+} \quad 24 \text{ MeV} \quad {^{4}\text{He}}^{+} \quad 1.7 \text{ MeV} \quad \text{(degraded with carbon foils)} \]

Dose \sim 30 \text{ dpa} \quad \text{[He]} \sim 25 \text{ appm/dpa}

A comparison of a steel with nano-particles (right) and a steel without the nano-particles (left).

The specimen with particles exhibits small bubbles all below the critical size. The specimen without particles shows the start of void growth.

EXAMPLE 4: ODS (OXIDE DISPERSED STRENGTHENED) STEELS

M. J. Fluss, S. Tumey, L. Hsiung (LLNL) and A. Kimura (Kyoto University)

Dual beam irradiation, details

Oxides nano-particles (labeled A thru F)

By varying the focusing condition on the right we see the small He bubbles (red arrows) trapped at the interface between the particles and the steel matrix. Oxide nanoparticles play the role of defect sinks by trapping He and preventing swelling.
EXAMPLE 4: ODS (OXIDE DISPERSED STRENGTHENED) STEELS

M. J. Fluss, S. Tumey, L. Hsiung (LLNL) and A. Kimura (Kyoto University)

ODS (K3 and MA957), Fe and Fe-Cr 14%

Triple beam irradiation

\[ ^{56}\text{Fe}^{8+} \rightarrow 24 \text{ MeV} \]

Dose \(\sim 30\) dpa

\[ ^{4}\text{He}^{+} \rightarrow 1.7 \text{ MeV} \text{ (degraded with carbon foils)} \]

[H] \(\sim 70\) appm/dpa

\[ T = 425^\circ\text{C} \]

The cavities exhibit a double shell structure

EXAMPLE 4: ODS (OXIDE DISPERSED STRENGTHENED) STEELS

K3-ODS (3-beam)

Fresnel fringes

5 nm

Defocus: -150 nm

Ce

Voids

5 nm

Defocus: -200 nm
Summary

Electron and ion irradiation experiments

- Electron: 0.15 – 2.5 MeV
- Ions: H to U, 0.3 – 1000 MeV

Combined with in situ or ex situ instrumentations:

- Low to high temperature: 8 K – 1200 K
- X-ray diffractometer
- IR spectrometer
- Gas analyser
- IBA
- UV-visible
- Electrical measurements
- Raman spectrometer
- TEM

Next call: October 2013
for the period of April 2014 to March 2015
http://emir.in2p3.fr/EMIR-network
Next EMIR User days in 2013
Visit of JANNUS-Saclay on Friday

Acknowledgments to the contributors
<table>
<thead>
<tr>
<th>Samples</th>
<th>Single crystal 6H-SiC</th>
<th>Single crystal TiC</th>
<th>Single crystal ZrC</th>
<th>Single crystal 6H-SiC</th>
<th>Single crystal TiC</th>
<th>Single crystal ZrC</th>
</tr>
</thead>
<tbody>
<tr>
<td>Facility</td>
<td>JANNUS</td>
<td>JANNUS</td>
<td>JANNUS</td>
<td>GANIL-SME</td>
<td>GANIL-SME</td>
<td>GANIL-SME</td>
</tr>
<tr>
<td>Ion</td>
<td>Au⁺</td>
<td>Au⁺</td>
<td>Au⁺</td>
<td>Pb⁵³⁺</td>
<td>Pb⁵³⁺</td>
<td>Pb⁵³⁺</td>
</tr>
<tr>
<td>Energy (MeV)</td>
<td>1.2</td>
<td>1.2</td>
<td>1.2</td>
<td>956</td>
<td>956</td>
<td>956</td>
</tr>
<tr>
<td>Rp</td>
<td>194 nm</td>
<td>219 nm</td>
<td>217 nm</td>
<td>37.13 µm</td>
<td>28.2 µm</td>
<td>26.5 µm</td>
</tr>
<tr>
<td>dpa</td>
<td>0.05-40.25</td>
<td>0.066-55.4</td>
<td>0.1-83.5</td>
<td>0.0002-0.0013</td>
<td>0.0002-0.0012</td>
<td>0.0003-0.0025</td>
</tr>
<tr>
<td>(dE/dx)ₜ (keV/µm)</td>
<td>2728</td>
<td>2147</td>
<td>1689</td>
<td>33080</td>
<td>43580</td>
<td>47160</td>
</tr>
<tr>
<td>(dE/dx)ₜ (keV/µm)</td>
<td>4051</td>
<td>3636</td>
<td>3736</td>
<td>61.03</td>
<td>81.62</td>
<td>102</td>
</tr>
<tr>
<td>Fluences(ions/cm²)</td>
<td>10¹³-10¹⁶</td>
<td>10¹³-10¹⁶</td>
<td>10¹³-10¹⁶</td>
<td>10¹²-10¹³</td>
<td>10¹²-10¹³</td>
<td>10¹²-10¹³</td>
</tr>
</tbody>
</table>