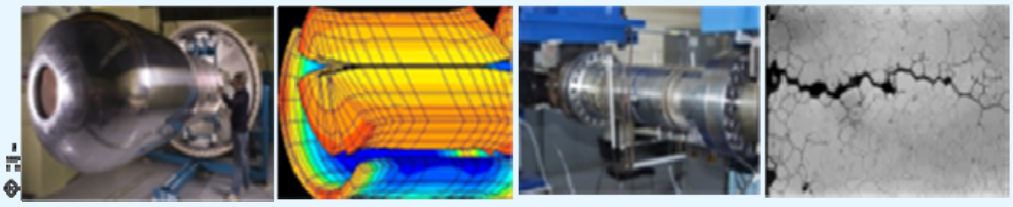
MINOS

Centre of Excellence for Nuclear Materials

Workshop

Materials Innovation for Nuclear Optimized Systems



December 5-7, 2012, CEA – INSTN Saclay, France

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

Lucile BECK[1]

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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).

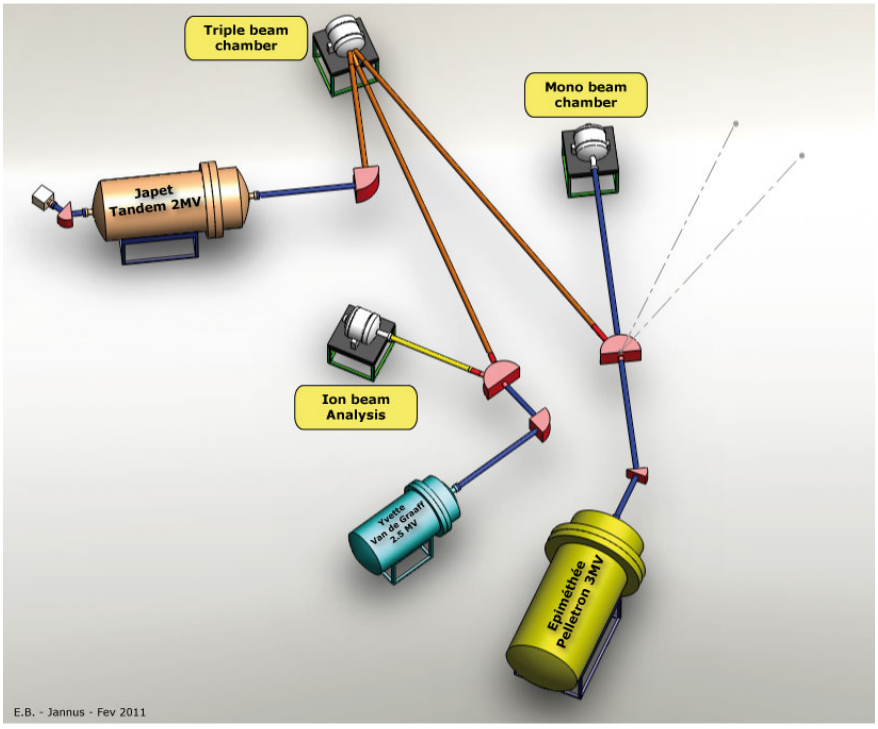

Fig. 1: Schematic view of the JANNUS platform at Saclay.

DE LA RECHERCHE À L'INDUSTRIE

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)

MINOS Workshop, Materials Innovation for Nuclear Optimized Systems
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PART 1 : CEA charged particle irradiation facilities

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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

within

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

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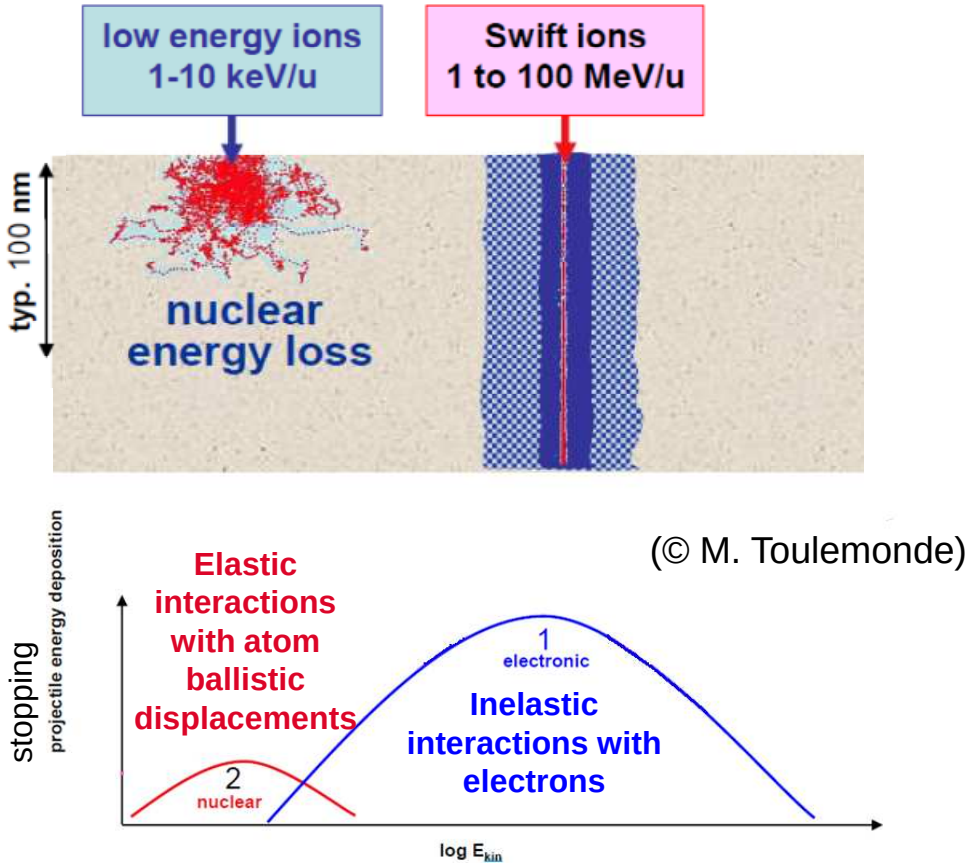
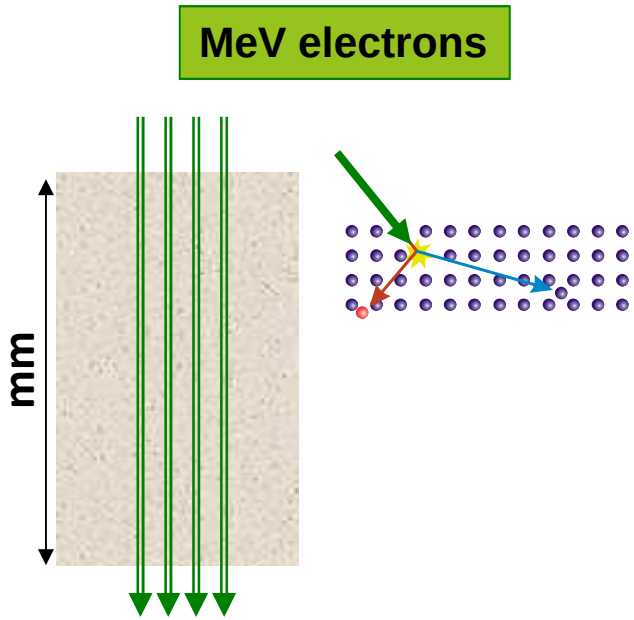
Scheme of energy deposition of an ion in matter / electrons in matter



(© M. Toulemonde)



Access to the CEA charged particle irradiation facilities

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emir facilities

ions

SRMP JANNUS-Saclay	Epiméthée	H → Bi	0.5 – 30 MeV
	Japet	H → Bi	12 MeV
	Yvette	H, ^{2}H, ^{3}He, ^{4}He	0.5 – 2.5 MeV

CSNSM JANNUS -Orsay	ARAMIS	H → Bi	0.5 – 15 MeV
	IRMA	H → Bi	5 – 570 keV

+ TEM

CIMAP	GANIL IRRSUD	C → U	0.3 – 1 MeV/A
	GANIL SME	C → U	4.5 – 13 MeV/A

electrons

LSI	SIRIUS	e-	0.15 – 2.5 MeV
SRMA	HVEM	e-	1.2 MeV

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

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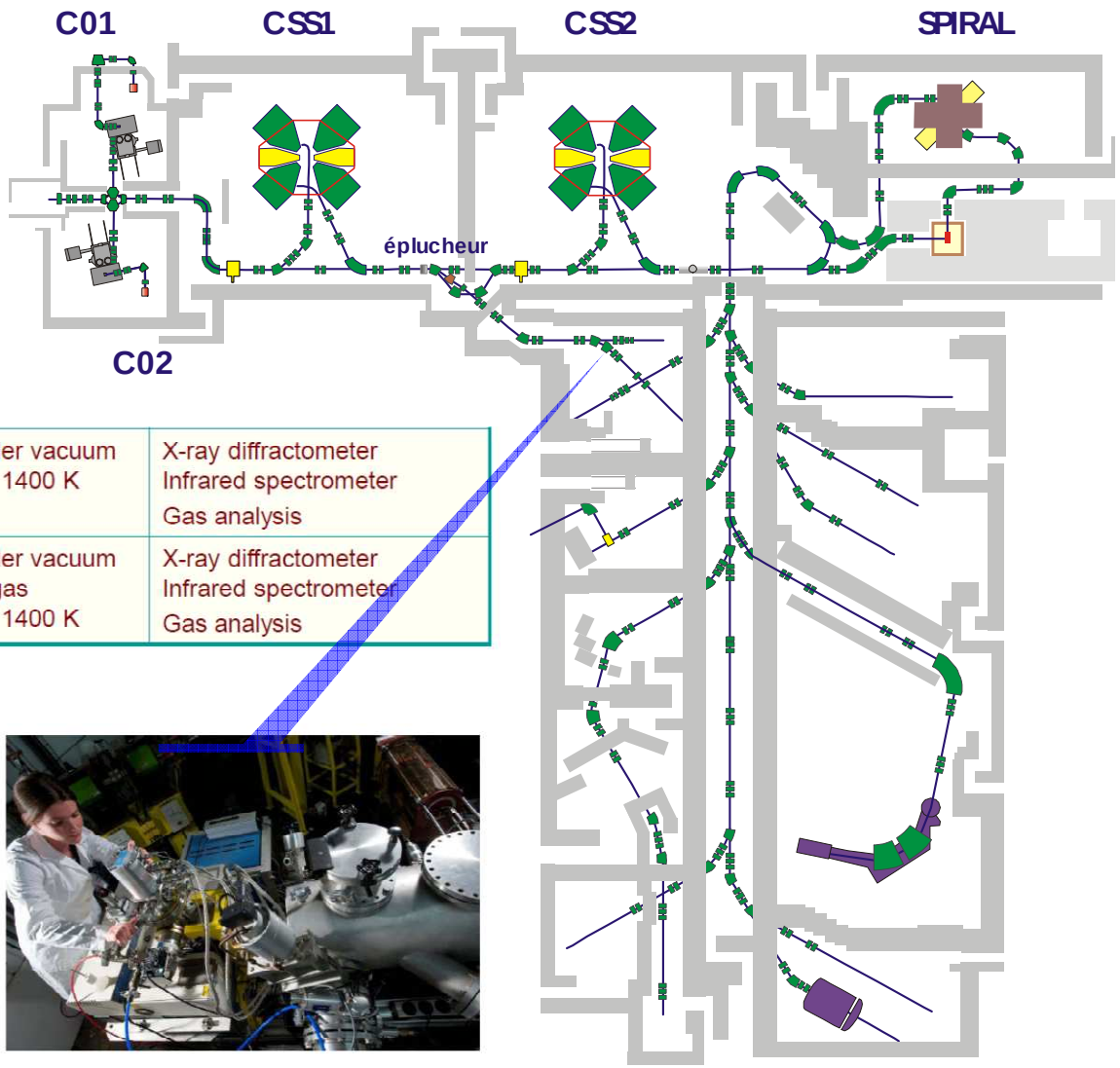
Cimap - Caen



GANIL IRRSUD	C → U	0.3 – 1 MeV/A	under vacuum 8 – 1400 K	X-ray diffractometer Infrared spectrometer Gas analysis
GANIL SME	C → U	4.5 – 13 MeV/A	under vacuum or gas 8 – 1400 K	X-ray diffractometer Infrared spectrometer Gas analysis

Scientific thematics

- *Physics of the ion – matter interaction*
- *Mechanisms of energy transfert,*
- *Damage mechanisms and structural modifications in materials,*
- *Simulation of the nuclear material behaviour under irradiation,*
- *Nanostructuration by irradiation,*
- *Radiation chemistry*
- *Radiobiology*

JANNUS-Saclay: 3 joint accelerators
Low to heavy ions – MeV energy range

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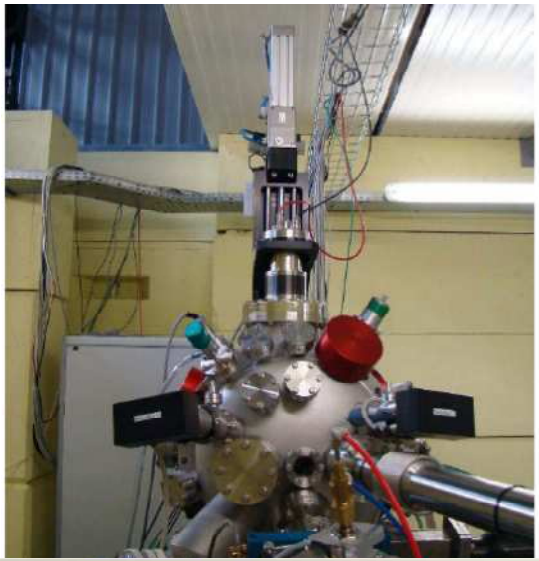
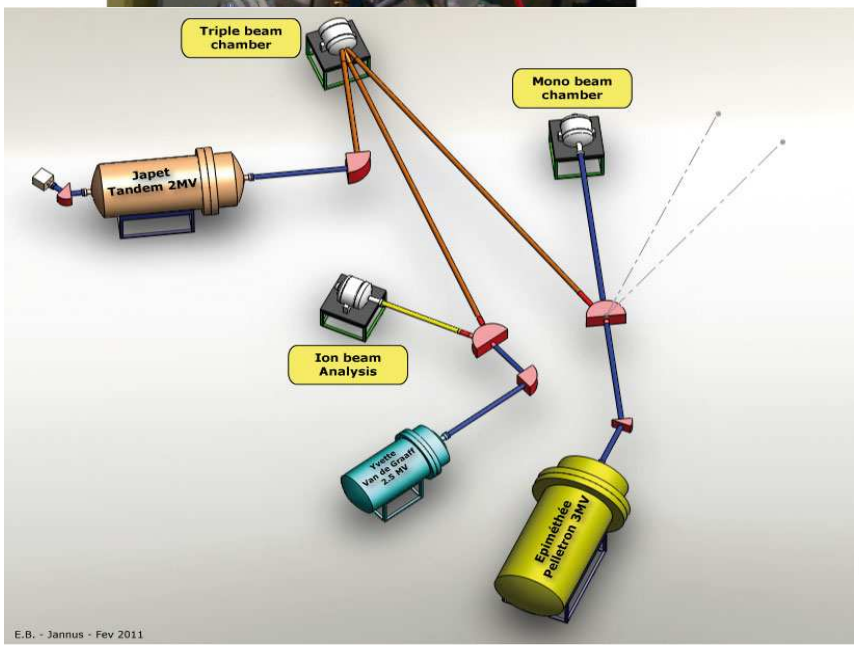


Epiméthée	H → Bi	0.5 – 30 MeV	under vacuum 77 – 800 K	
Japet	H → Bi	tandem 2 MV	under vacuum 77 – 800 K	
Yvette	H, ^{2}H, ^{3}He, ^{4}He	0.5 – 2.5 MeV	under vacuum	NRA – RBS ERDA – PIXE



Scientific thematics

- *Microstructure evolution under irradiation for nuclear materials*
- *Synergetic effects of damage and Helium and Hydogen production*
- *Atomic transport phenomena under irradiation*
- *Controled modification by irradiation of the material properties*
- *Teaching and formation with the accelerators*

JANNUS

LSI-Palaiseau: MeV energy electrons

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LSI - Palaiseau

SIRIUS

SIRIUS	e-	0.15 – 2.5 MeV	High doses (GGy) Large area 20 – 600 K	Optical spectroscopies Micro-electrochemistry Electrical measurements Gas emission

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*

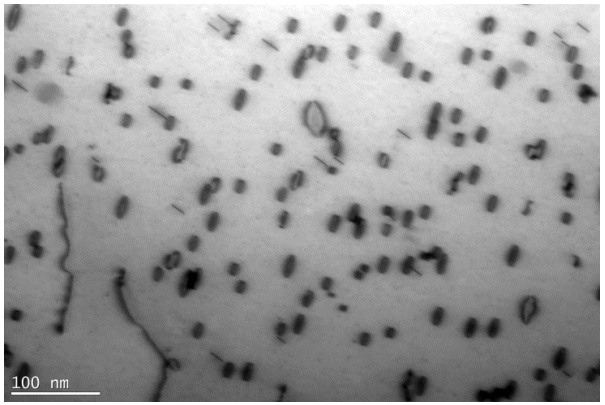
SRMA-Saclay: MeV energy electrons

SRMA - Saclay

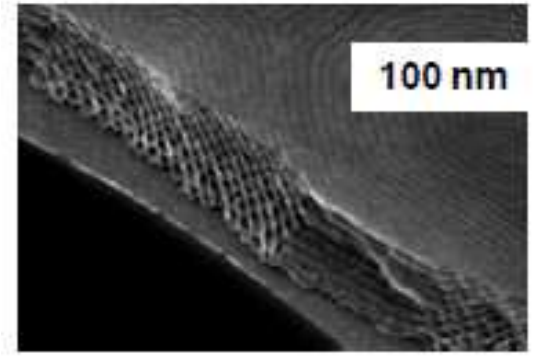
HVEM	e-	0.3 – 1.2 MeV	under vacuum	TEM

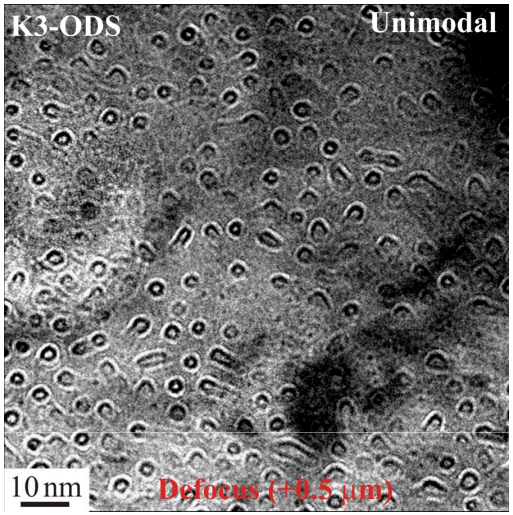
Scientific thematics

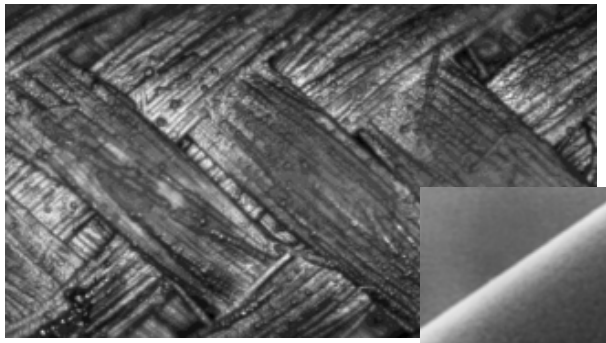
- *Microstructure evolution under irradiation for nuclear materials*
- *Effects of the high damage rate*

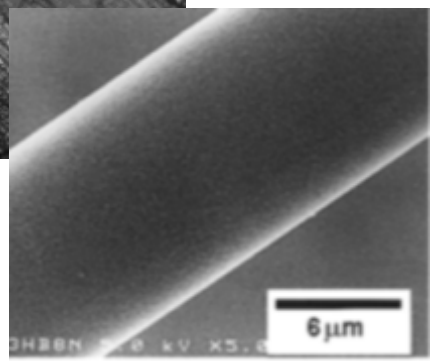


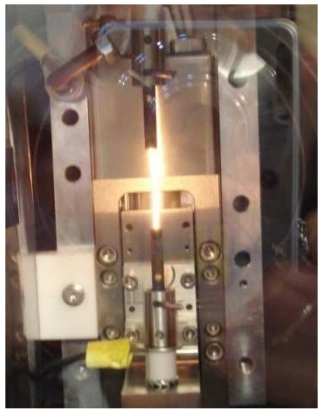
PART 2: examples

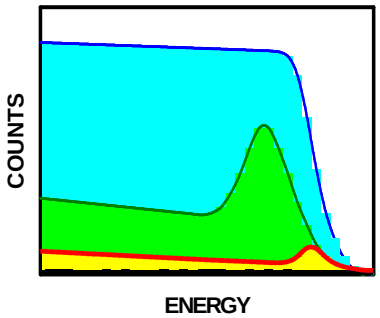












Example 1: Mesoporous silica gels for applications in the field of nuclear waste

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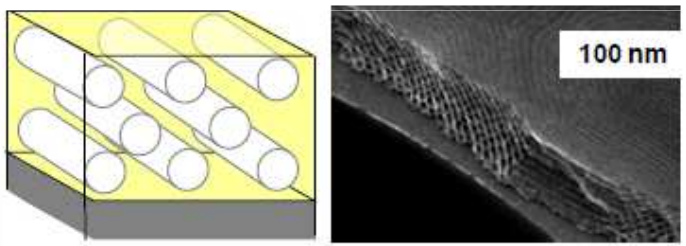
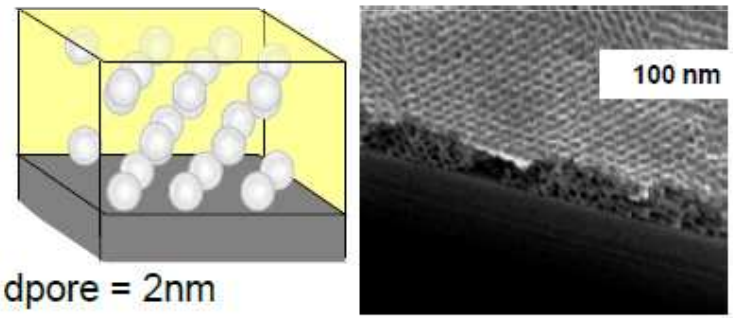
•Sample characteristics

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

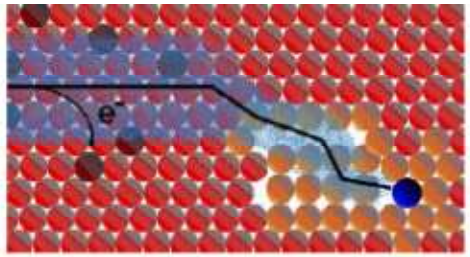
SBA15 : Structure 2D hexagonale	Structure 3D hexagonale
 dpore=5nm	 dpore = 2nm

Separation chemistry
Waste management

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

Example 1: Mesoporous SiO_2 gel

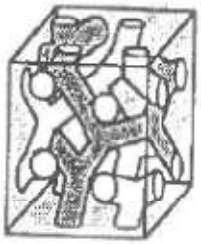
X. Deschanels, S. Dourdain and coll.
ICSM, Marcoule



Inelastic processes
Ganil, CEMHTi

Ballistic processes
Jannus

Mesporous material



Irradiations conditions

✓ Inelastic processes Xe : 92 MeV
-1x10^{13} cm^{-2} -$T_{amb.}$
$(dE/dx)_e$=9 keV/nm

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

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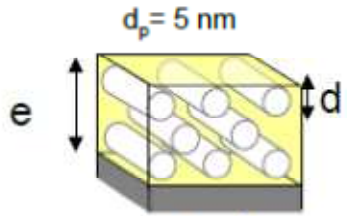
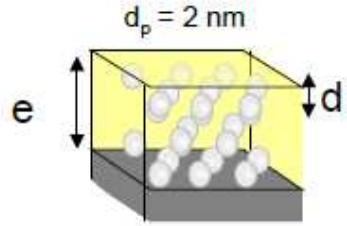


dpore=5nm empty

Xe : 92 MeV

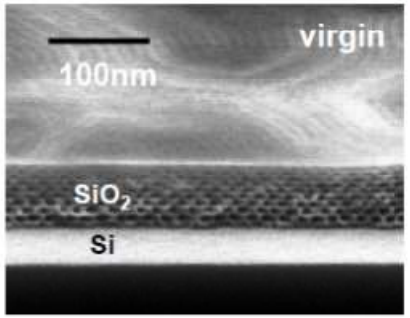
$1x10^{13}$ cm^{-2} -Tamb.

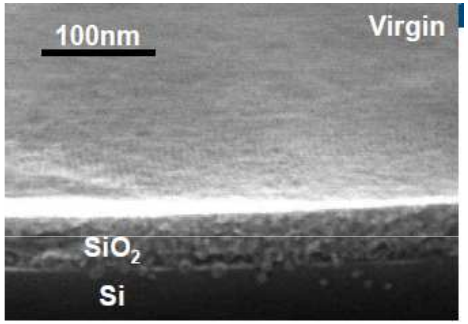
dE/dx=9keV/nm

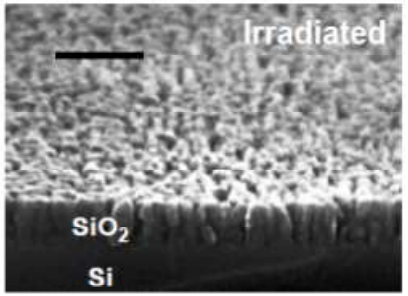


dpore = 2nm

@ GANIL



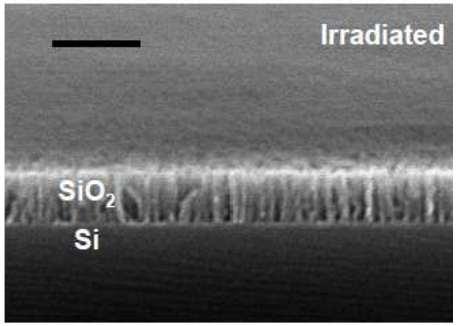




•Parameters of the coating

✓Thickness decrease

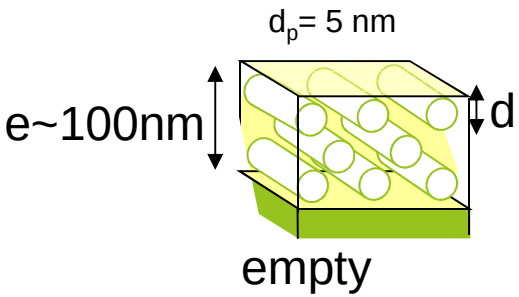
✓Porosity volume increase

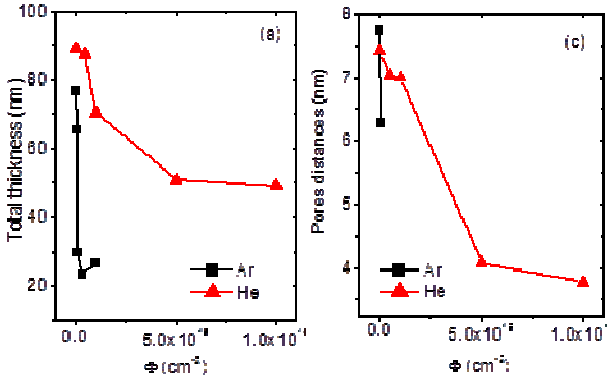


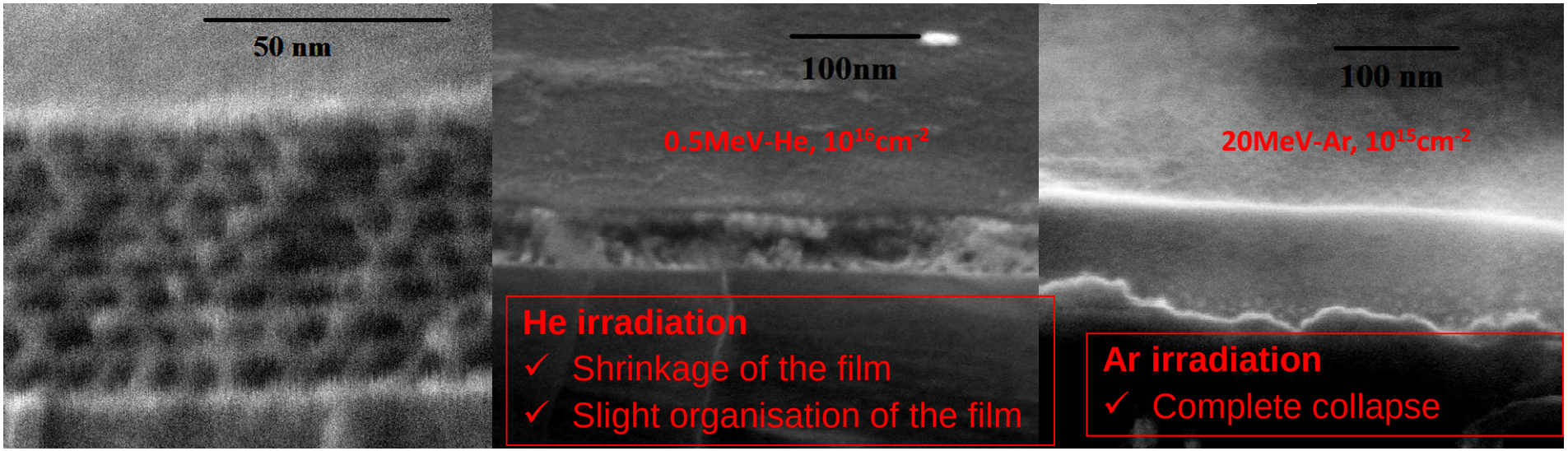
– Destructuration of the silica network under Xe 92MeV

Example 1: Mesoporous SiO_2 gel

0.5 MeV He, 20 MeV Ar @ JANNUS







He irradiation
- ✓ Shrinkage of the film
- ✓ Slight organisation of the film

Ar irradiation
- ✓ Complete collapse

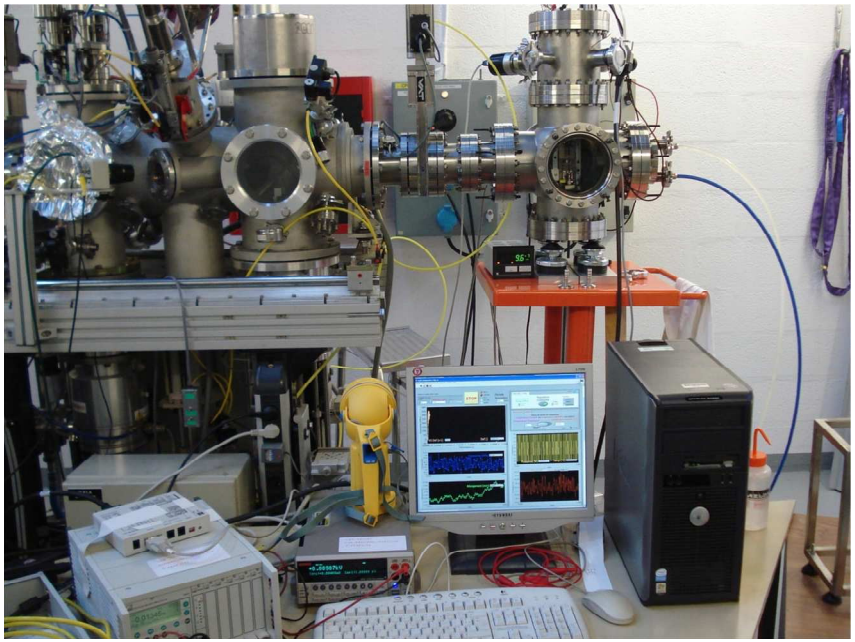
Densification and loss in the organisation of the thin films even for S_e lower than track threshold

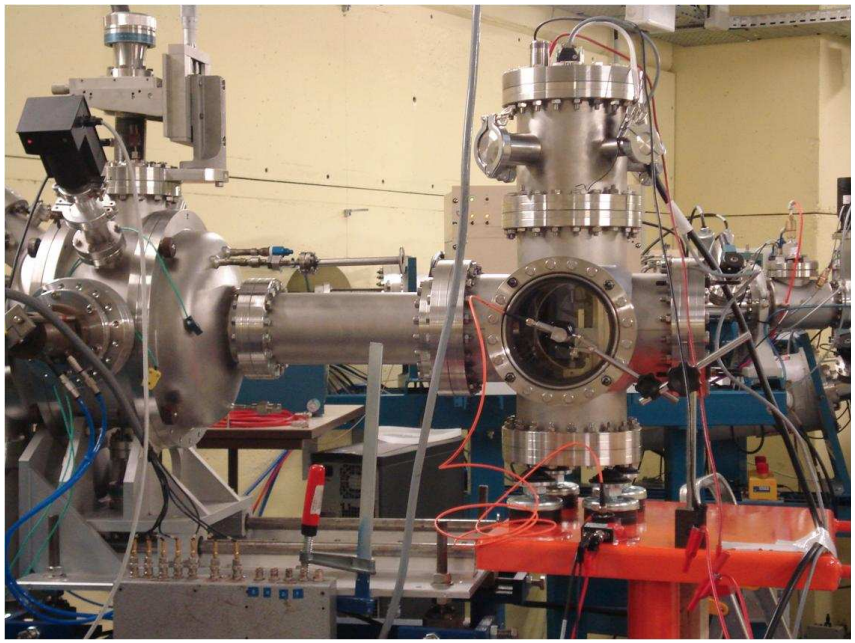
Example 2: *In-situ* mechanical test

IRRSUD line GANIL Caen

Épiméthée E3 line JANNUS Saclay

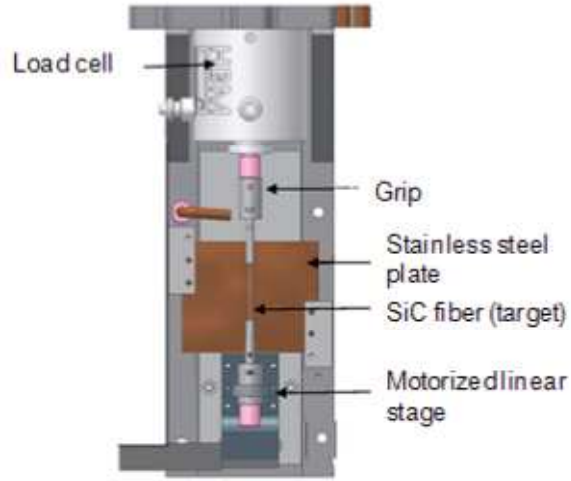
⇒ Damage effect for both electronic and nuclear slowing down regimes

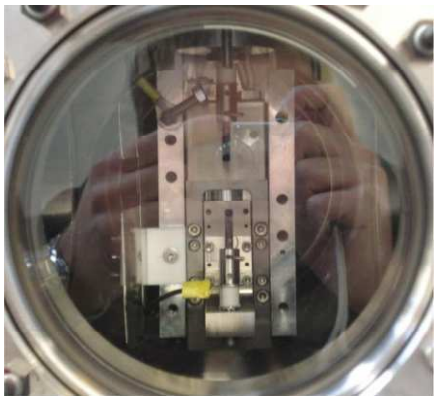


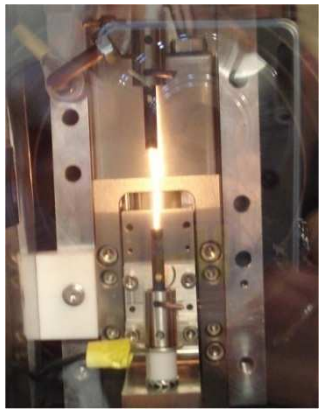


A. Jankowiak, Ch. Colin (CEA/DM//SRMA) et coll.

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

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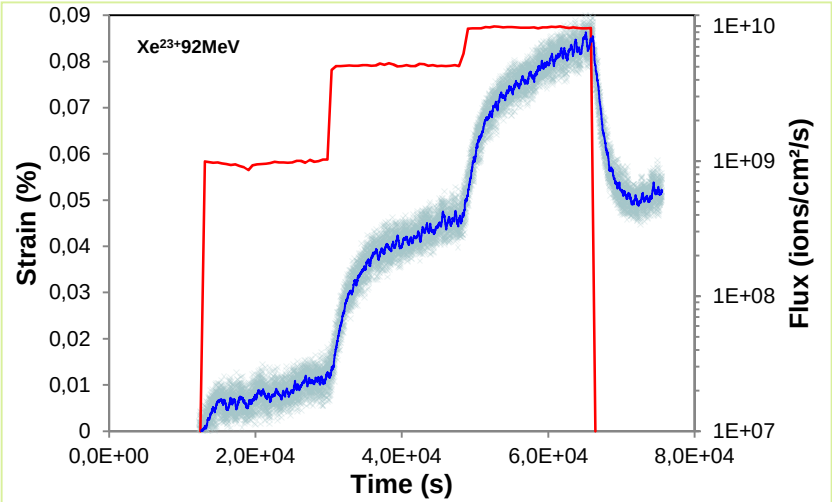
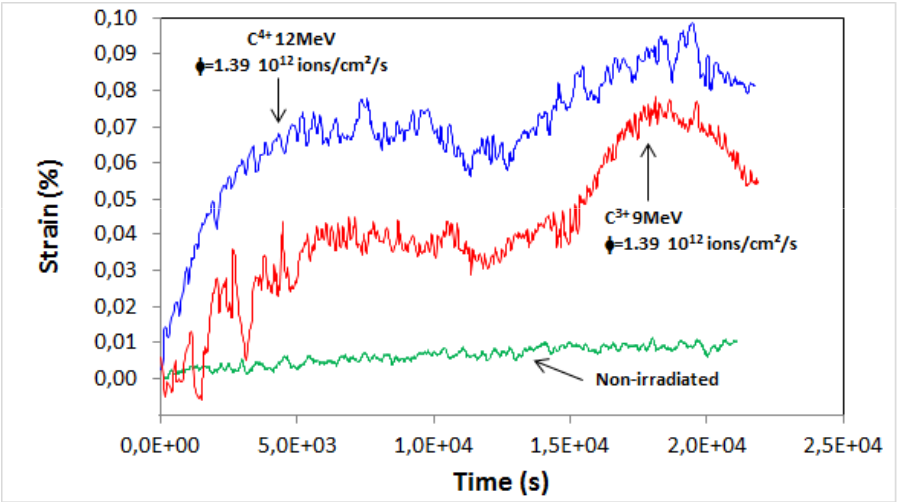
In situ tensile tests on SiC fibers under ion beam

IRRSUD line GANIL Caen **Xe^{23+} 92 MeV**

Épiméthée E3 line JANNUS Saclay **C^{+} 9 and 12 MeV**

Fiber at 1000 ℃ and 300MPa





Fiber at 1000 ℃ and 300MPa (irradiation-enhanced c reep study)

- Strain rate variation with flux intensity;
- Residual stain at 1000 ℃ < 0.1 % for 2.10^{12} 92 MeV Xe^{23+} ions/cm^2 and 3.10^{16} 12 MeV C^{4+} ions/cm^2

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

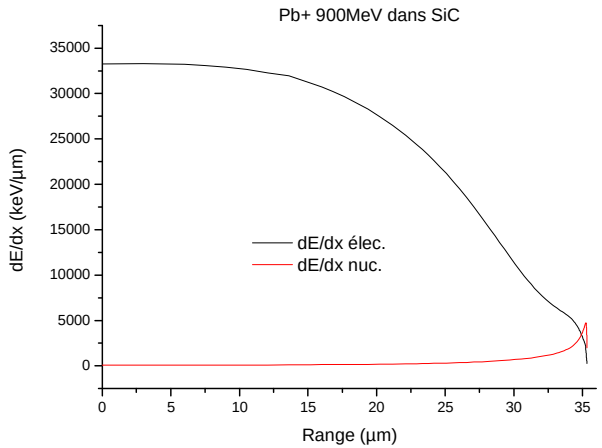
- Residual strain at RT ~ 0,50% % for 5.10^{14} 92 MeV Xe^{23+} ions/cm^2 and ~0.45 % for 10^{17} 12 MeV C^{4+} ions/cm^2
- Annealing test at 1800℃: residual strain almost disapp ears completely

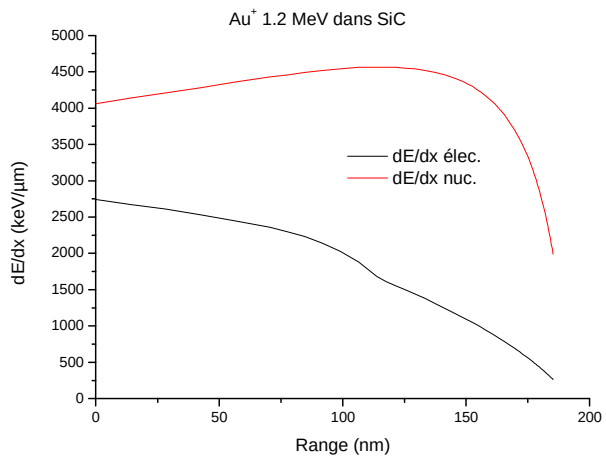
Example 3: STUDY OF RADIATION EFFECTS IN CARBIDES (SiC, TiC, ZrC)

PhD thesis of S. Pellegrino (JANNUS): "*Irradiation effects in carbides (SiC, TiC and ZrC) and synergetic effects of electronic and nuclear energy losses.*"

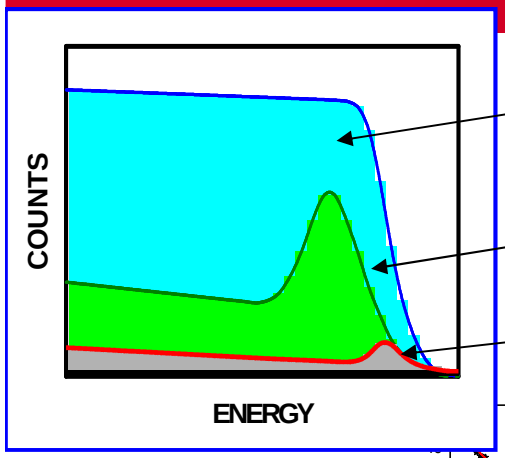
GANIL-SME
Pb 900 MeV
High Energy
$(dE/dx)_{nucl.} << (dE/dx)_{elec.}$

JANNUS
Au 1,2 MeV
Low Energy
$(dE/dx)_{nucl.} > (dE/dx)_{elec.}$





DAMAGE CHARACTERIZATION WITH RBS in CHANNELING conditions (CSNSM-Orsay)



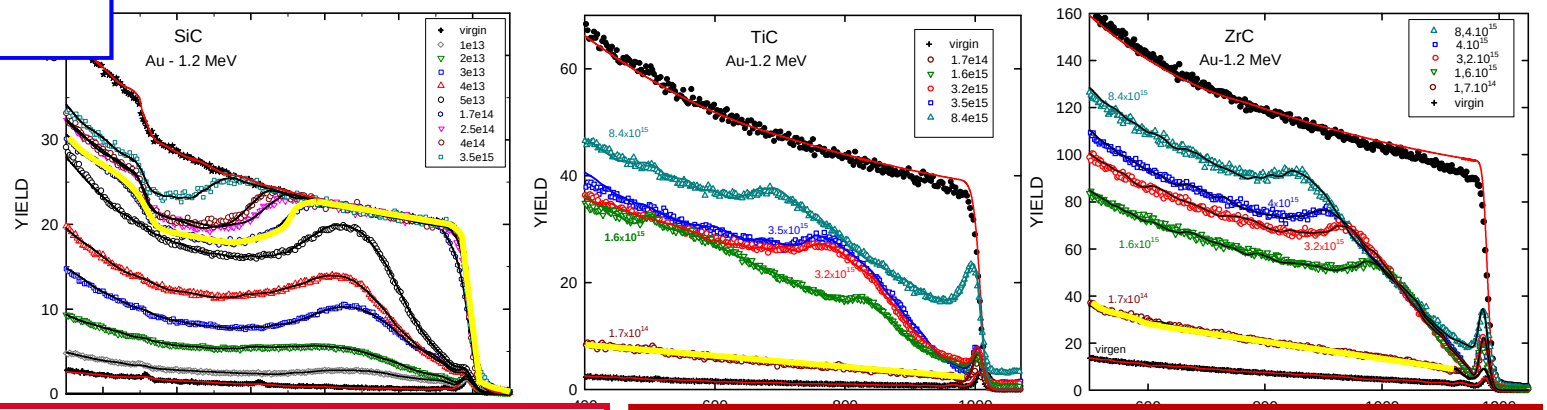
Amorphization
(single crystal in random orientation)

single crystal with damage

single crystal (oriented)

Damage

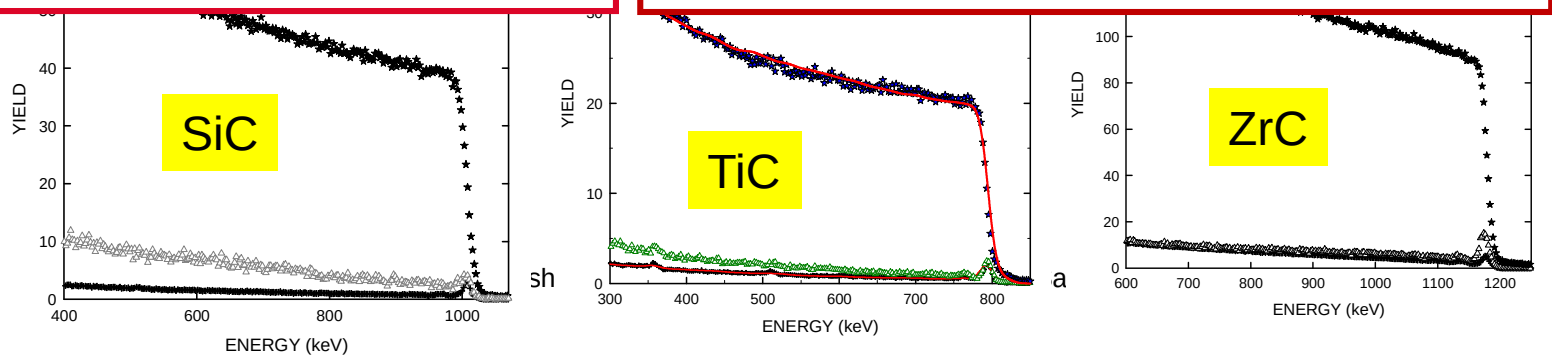
Au 1.2 MeV



Weak effect of electronic energy loss in SiC

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

Pb 900 MeV



EXAMPLE 4: ODS (OXIDE DISPERSED STRENGTHENED) STEELS

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

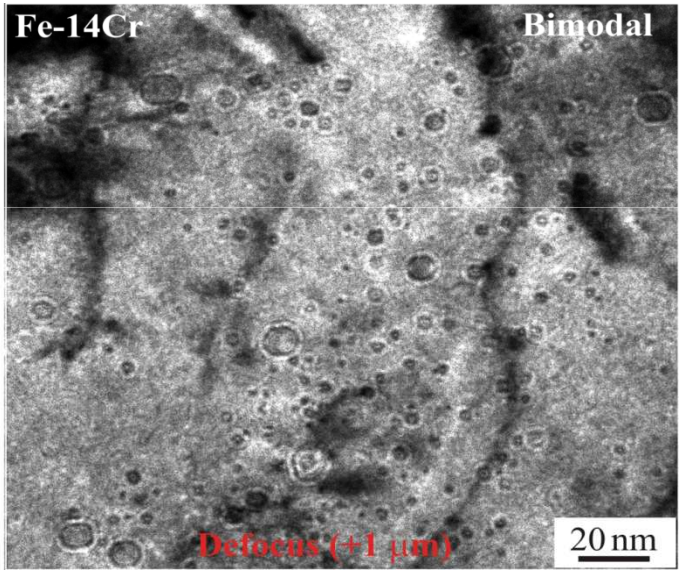
MINOS

Dual beam irradiation

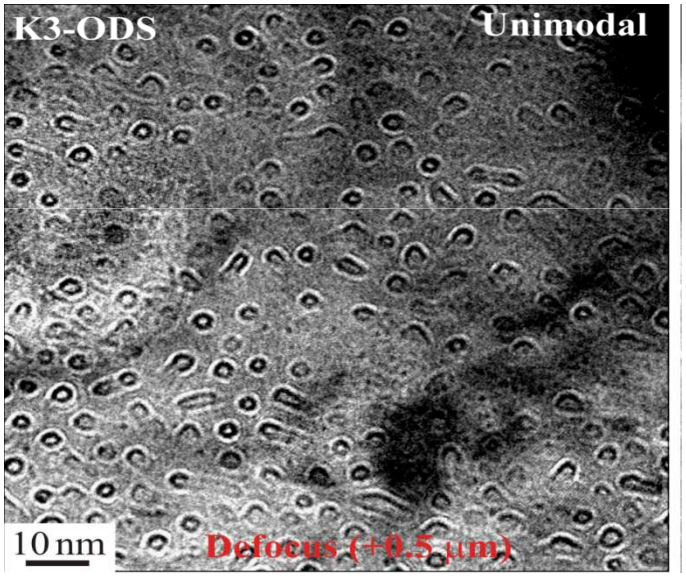
ODS (K3 and MA957) Fe-Cr 14%

$^{56}Fe^{8+}$ 24 MeV $^{4}He^{+}$ 1.7 MeV (degraded with carbon foils)

Dose ~ 30 dpa [He] ~ 25 appm/dpa



Start of void growth. Swelling



Small bubbles all below the critical size. No swelling

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.

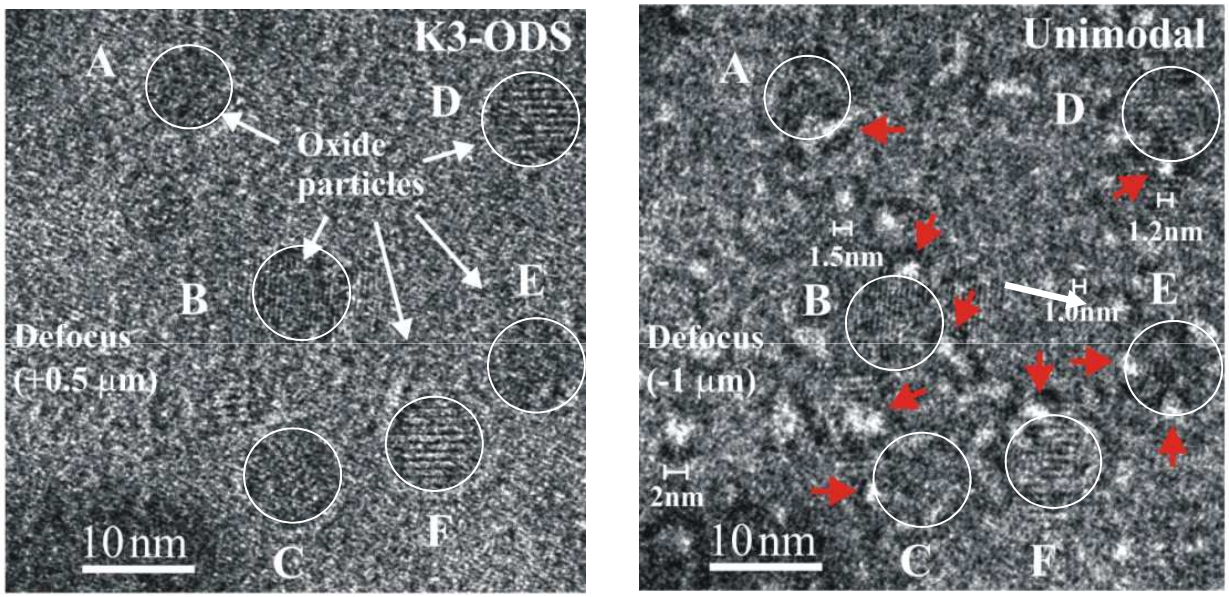
L. Hsiung, M. Fluss et al. (LLNL & SRMP), Phys. Rev. B 82, 2010, 184103

EXAMPLE 4: ODS (OXIDE DISPERSED STRENGTHENED) STEELS

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

MINOS

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)

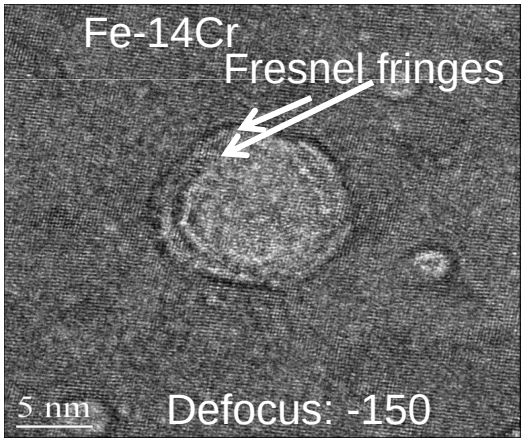
MINOS

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

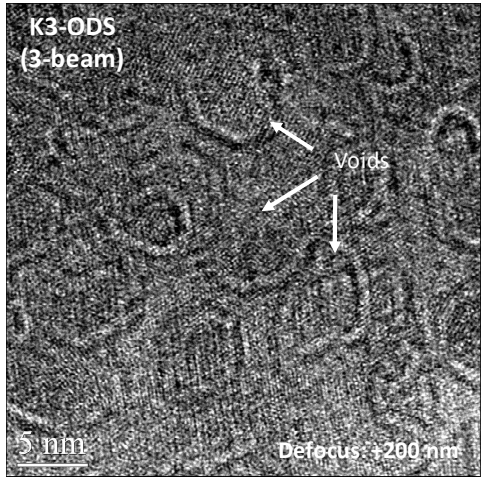
Triple beam irradiation

$^{56}Fe^{8+}$	$^{4}He^{+}$	H^{+}
24 MeV	1.7 MeV (degraded with carbon foils)	562 keV (degraded with carbon foils)
Dose ~ 30 dpa	[He] ~ 25 appm/dpa	[H] ~ 70 appm/dpa

T = 425℃



The cavities exhibit a double shell structure



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

Samples	Single crystal 6H-SiC	Single crystal TiC	Single crystal ZrC	Single crystal 6H-SiC	Single crystal TiC	Single crystal ZrC
Facility	JANNUS	JANNUS	JANNUS	GANIL-SME	GANIL-SME	GANIL-SME
Ion	Au^{+}	Au^{+}	Au^{+}	Pb^{53+}	Pb^{53+}	Pb^{53+}
Energy (MeV)	1.2	1.2	1.2	956	956	956
Projected Range: R_p	194 nm	219 nm	217 nm	37.13 µm	28.2 µm	26.5 µm
dpa	0.05-40.25	0.066-55.4	0.1-83.5	0.0002-0.0013	0.0002-0.0012	0.0003-0.0025
$(dE/dx)_e$ (keV/µm)	2728	2147	1689	33080	43580	47160
$(dE/dx)_n$ (keV/µm)	4051	3636	3736	61.03	81.62	102
Fluences(ions/cm²)	10^{13}-10^{16}	10^{13}-10^{16}	10^{13}-10^{16}	10^{12}-10^{13}	10^{12}-10^{13}	10^{12}-10^{13}