Behaviour of mesoporous silica under ion irradiation

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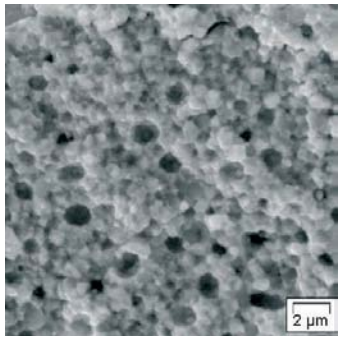
Mesoporous material in nuclear industry

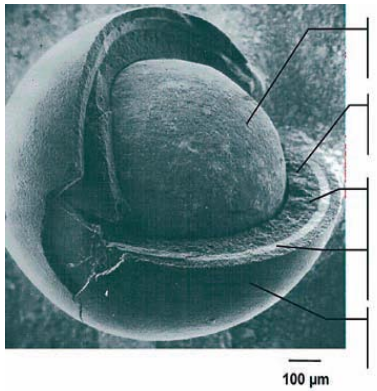
- Fuel forms

« RIM effect »



RHT fuel « Triso coated particles »

Fission products encapsulation in the porous strudture



UO_2 fuel kernel

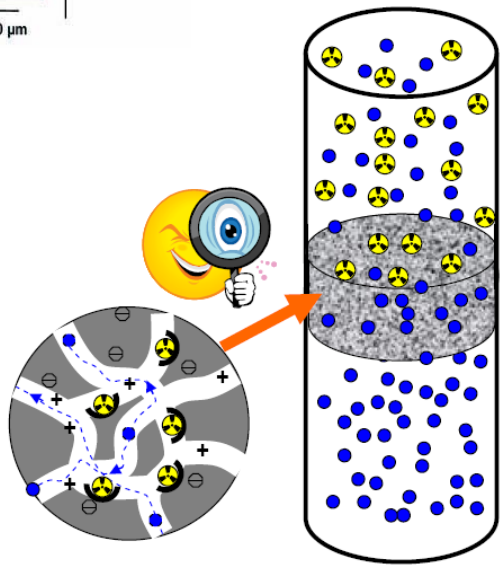
Porous carbon layer

Inner pyrolitic carbon layer

SiC layer

Outer porous carbon layer

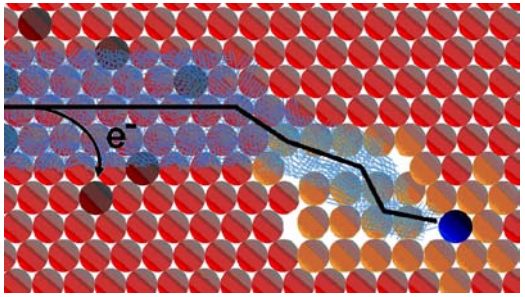
- Separation chemistry
  - Mesoporous SiO_2 for actinide extraction in from HLW
  - Mesoporous TiO2 as « getters materials » in geological repositories
  - Mesoporous ceramics for decontamination of liquid



- Waste management
  - Adsorption of the selected radionuclide
  - Encapsulation of the radionuclide by subsequent collapse of the structure (Thermal stress, chemical stress…)

A. Grandjean, X. Deschanels and al., ”Mesoporous materials in the field of nuclear industry: Applications and perspectives” submitted to NJC

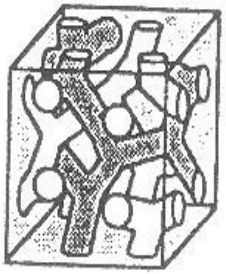
How irradiation modify the mesoporous structure?



Inelastic processes
Ganil, CEMHTi

Ballistic processes
Jannus

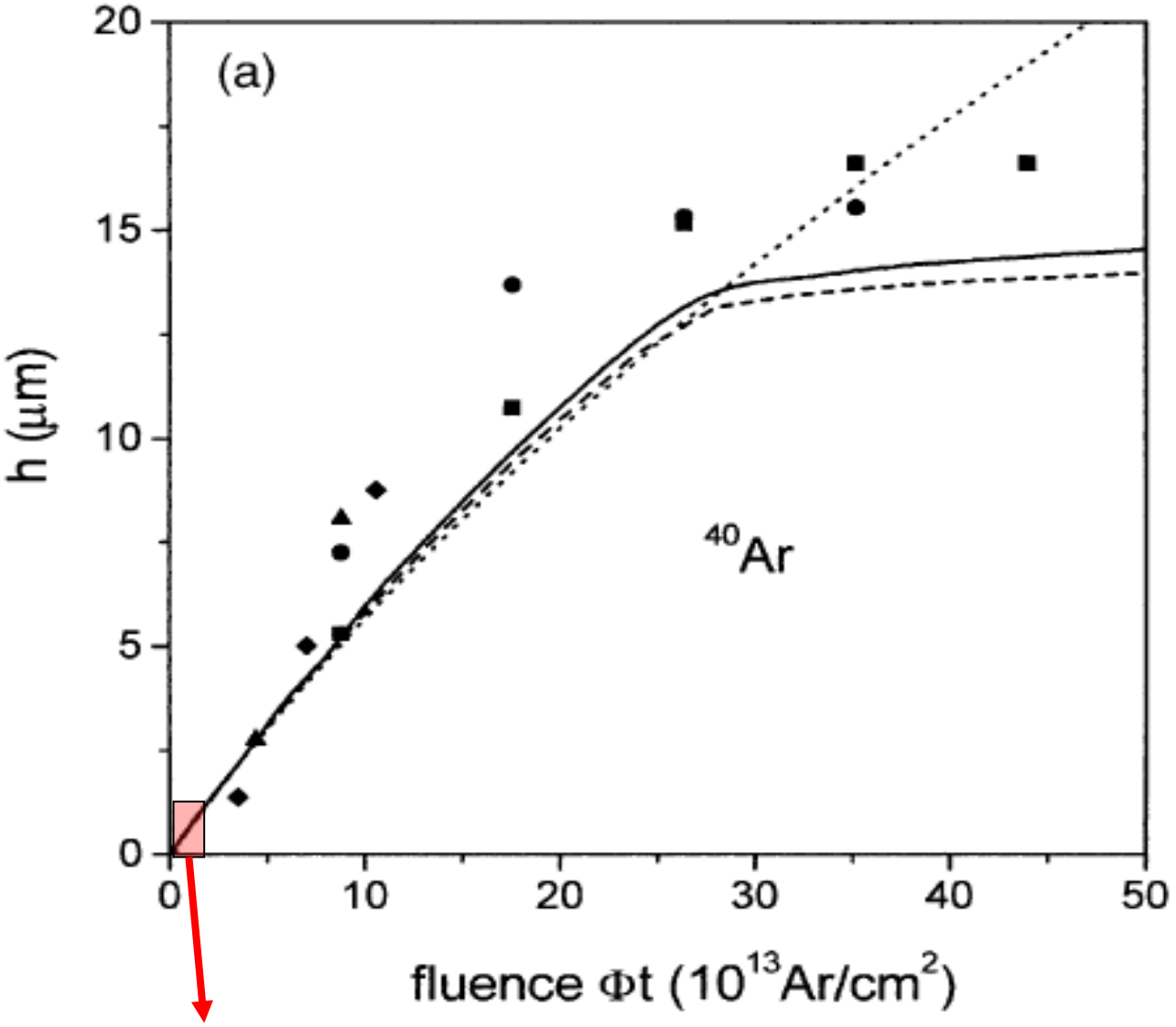
Mesporous material



- ✓Materials
  - •Vycor glasses (e~200µm)
  - •Mesoporous SiO_2 gels (e~100nm)
- ✓Analysis
  - •**BET, SAXS, X-Rays reflectometry**
  - MET, SEM

- ✓Materials
  - •Mesoporous SiO_2 gels (e~100nm)
- ✓Analysis
  - •X-Rays reflectometry
  - •MET, SEM

Bibliography



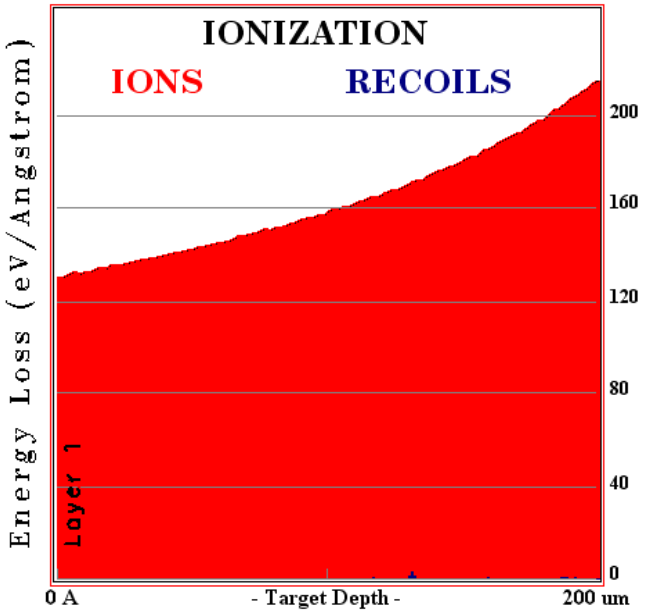
viscous flow

Fluence investigated in this study

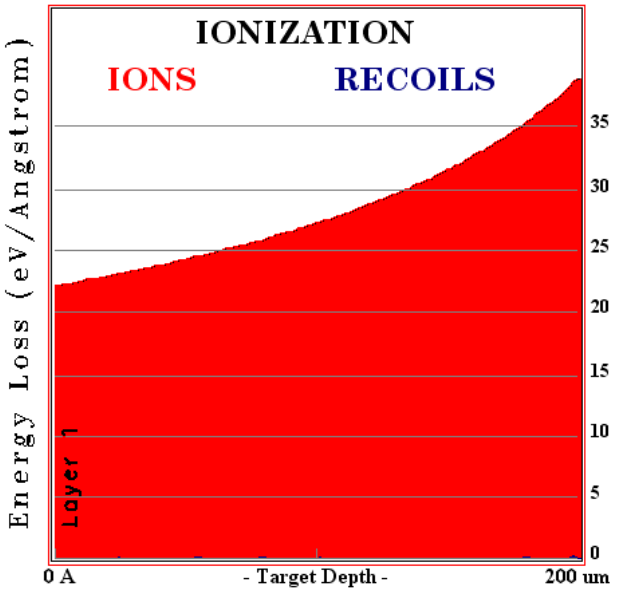
S.O. Kucheyev and al., «Light-ion-irradiation-induced thermal spikes in nanoporous SiO_2 » J. Phys. D : Appl. Phys. 44 (2011)

Irradiation – Mesoporous Vycor glasses

- Platelets 1x0.8x0.018cm^3 (design for BET measurements)
- Some samples filled with C and Cs to simulate a radionuclide
- Pore size : 3 nm – Pore surface : 125 m2/g
- Irradiations conditions
  - H : 17MeV / 0.05-0.1-0.5-1x10^{15} cm^{-2} ($T_{amb.}$) - $(dE/dx)_e$=0.0071 keV/nm
  - He : 32MeV / 0.05-0.1-0.5-1x10^{15} cm^{-2} ($T_{amb.}$) - $(dE/dx)_e$=0.03 keV/nm
  - C : 164 MeV / 0.3-0.5-1-3x10^{13} cm^{-2} ($T_{amb.}$) - $(dE/dx)_e$=0.18 keV/nm
  - Ar : 245 MeV / 0.3-0.5-0.8-1x10^{13} cm^{-2} ($T_{amb.}$) - $(dE/dx)_e$=2.3 keV/nm
  - Xe : 92 MeV / 1x10^{13} cm^{-2} ($T_{amb.}$) - $(dE/dx)_e$=9 keV/nm



C-100 MeV in SiO_2 (ρ=1,6g/cm^3)



Ar-500 MeV in SiO_2 (ρ=1,6g/cm^3)

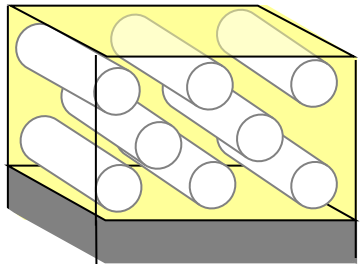
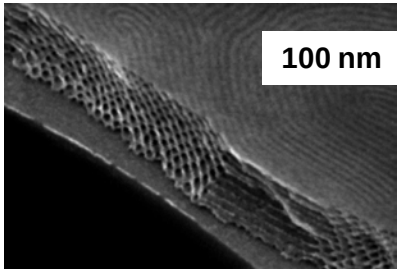
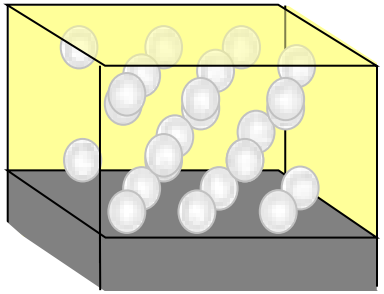
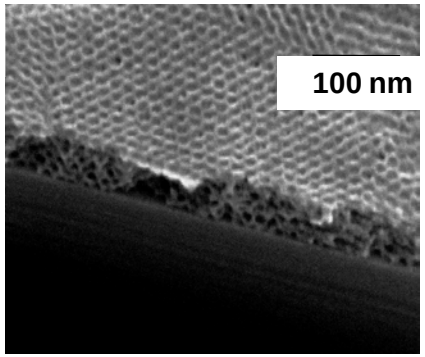
Irradiation – Mesoporous SiO_2 gels

•Sample characteristics

–Si substrate

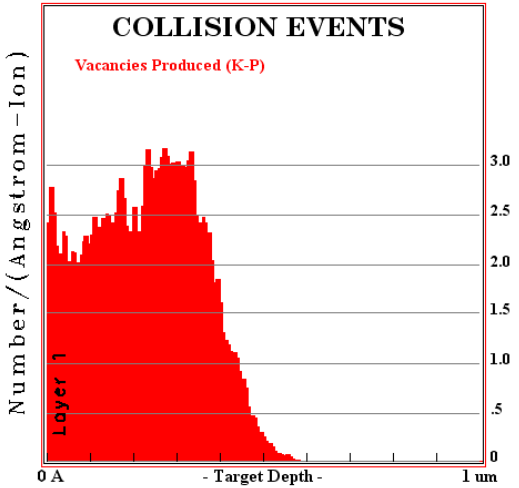
–Some samples filled with C and Cs to simulate a radionuclide

–Layer thickness : ~100nm

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

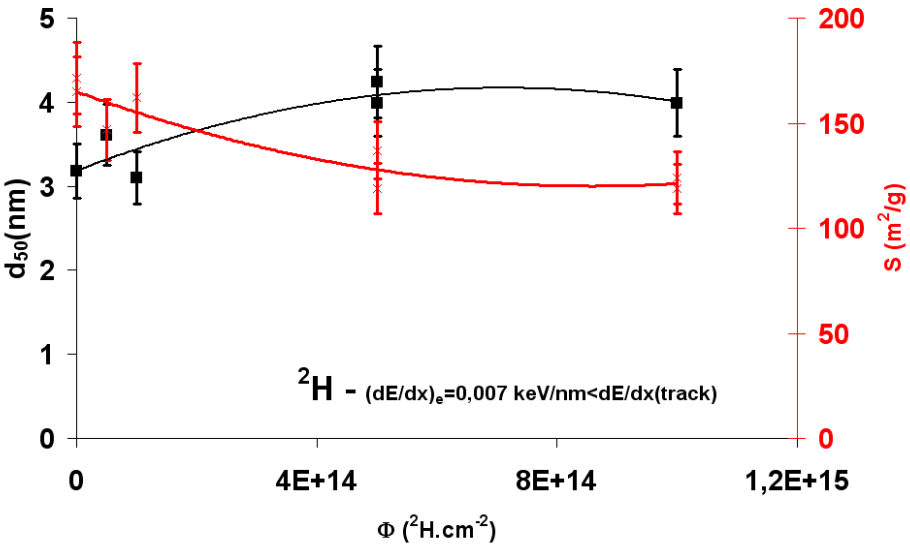
Irradiations conditions

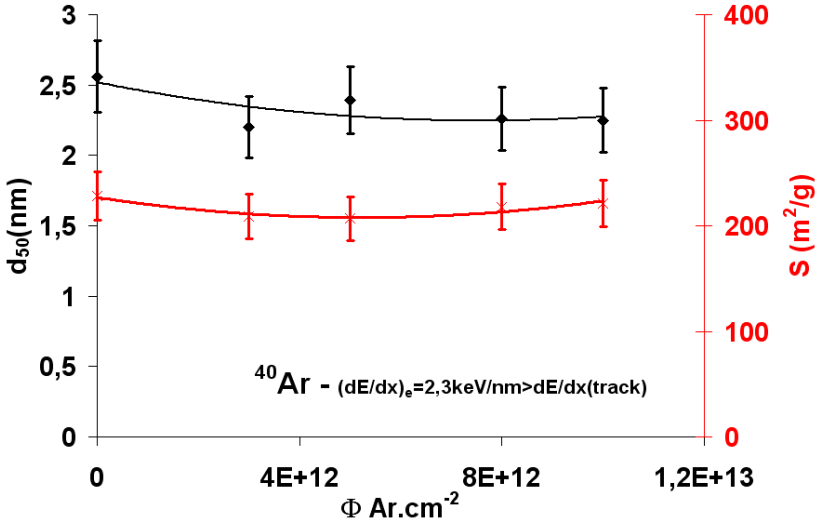
✓ Inelastic processes Xe : 92 MeV
-$1x10^{13}$ cm^{-2} -$T_{amb.}$



Au-1.5 MeV in SiO_2 (SRIM)

Preliminary results - BET measurements on Vycor glass

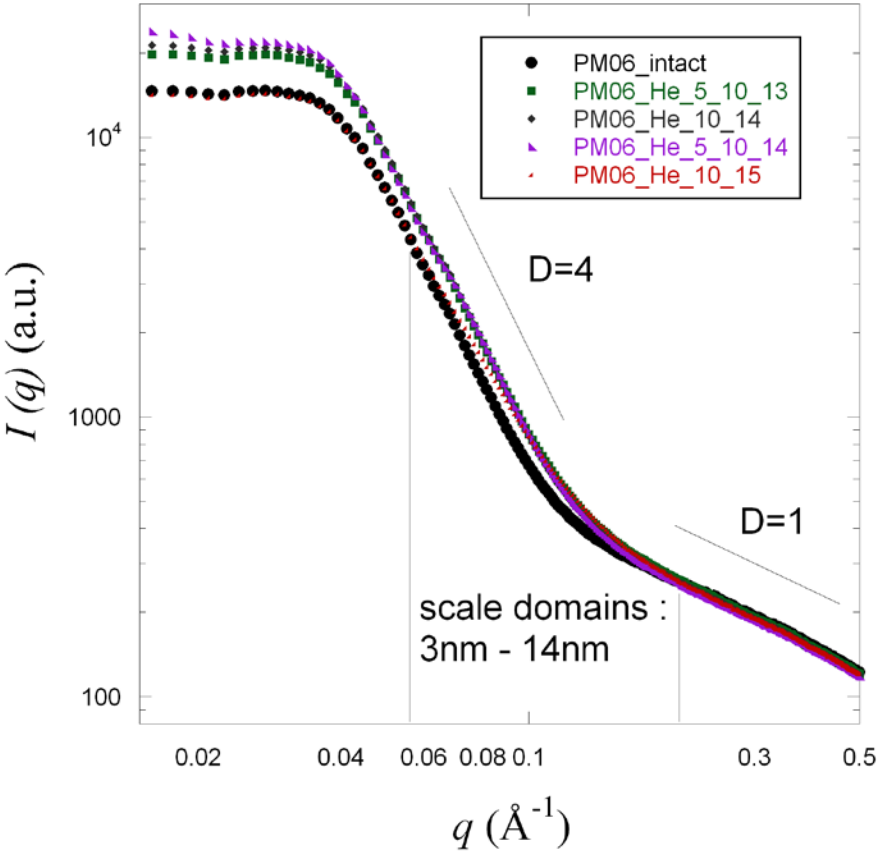


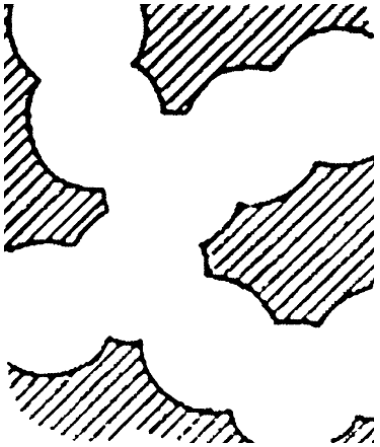


- Decrease of the surface area of the samples under irradiation
- Pore diameter
  - Small increase with deutons, He irradiations
  - Small decrease with C and Ar irradiations

Greater effects are expected for higher fluences

Preliminary results - SAXS measurements on Vycor glass





Schematic representation of virgin sample

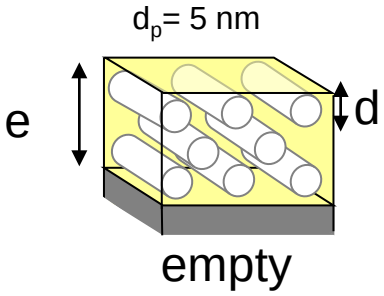
Increase of intensity at low *q* of samples from 5.10^{13} to 5.10^{14} MeV

Increase of volume fraction of pores ? increase of pore sizes ?

Change of fractal domain *D* at maximum fluence (10^{15} He/cm^2) : D<4

Appearance of rugosity of interfaces (pore surface)

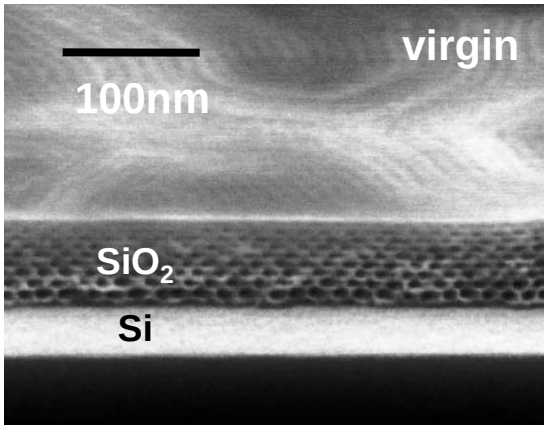
Irradiation – Mesoporous silica SBA 15

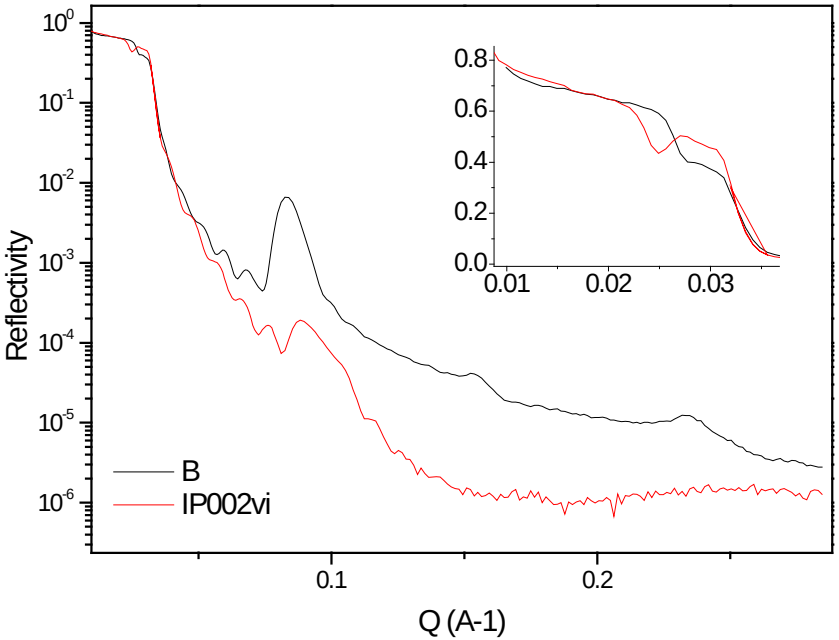


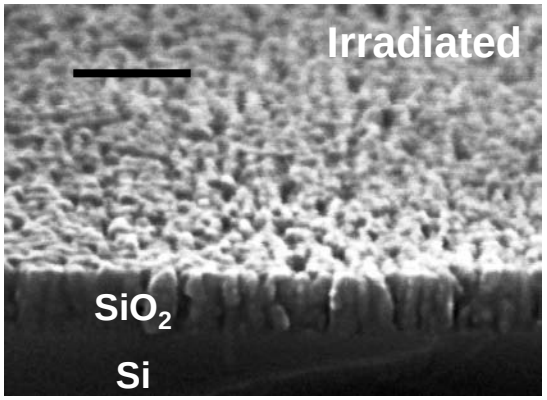
Xe : 92 MeV

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

dE/dx=9keV/nm





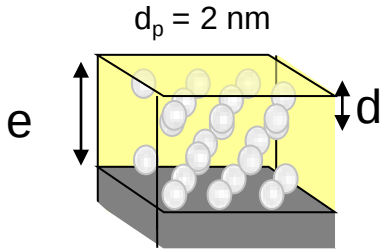


•Parameters of the coating

- **✓Thickness decrease (88 to 68 nm)**
- **✓Number of porous layers 10 to 9**
- **✓Pore diameter increase (5.8 to 7.3nm)**
- **✓Porosity volume increase 0.47 à 0.38 e-/Å^3**

Tracks formation

Irradiation – Mesoporous SiO_2 3D hex



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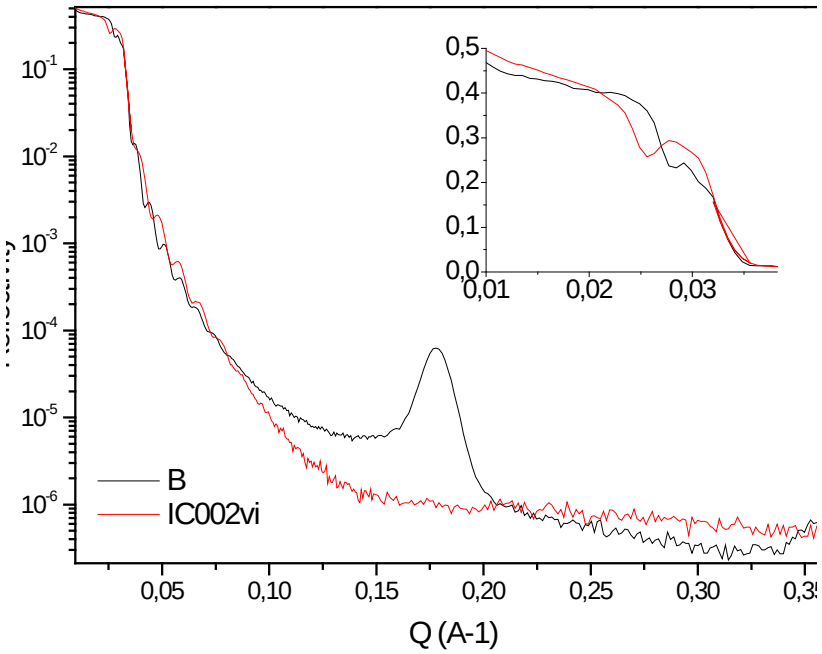
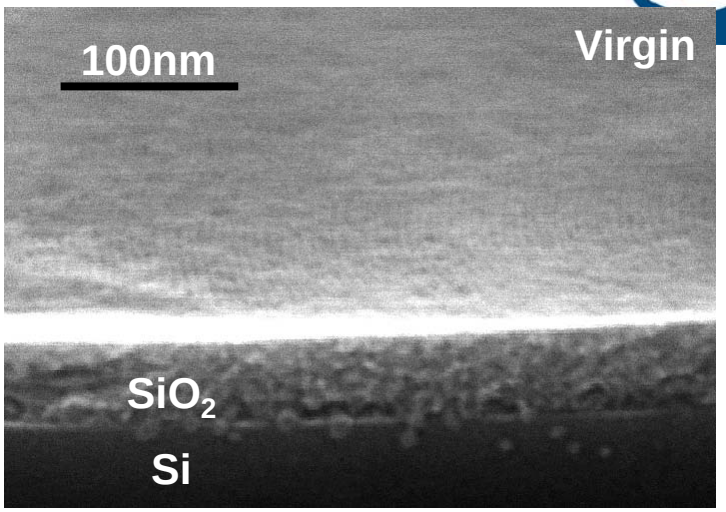
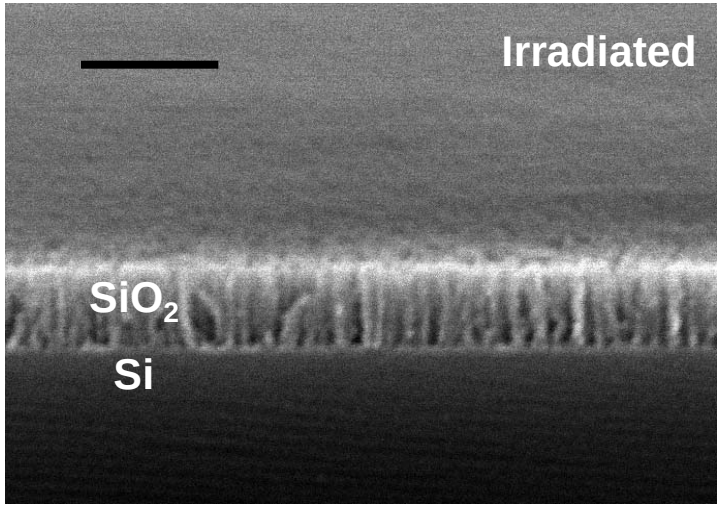
dE/dx=9keV/nm





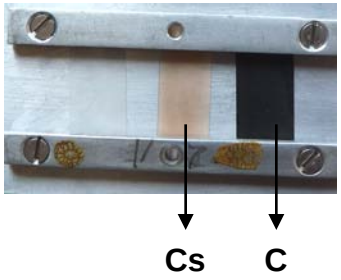


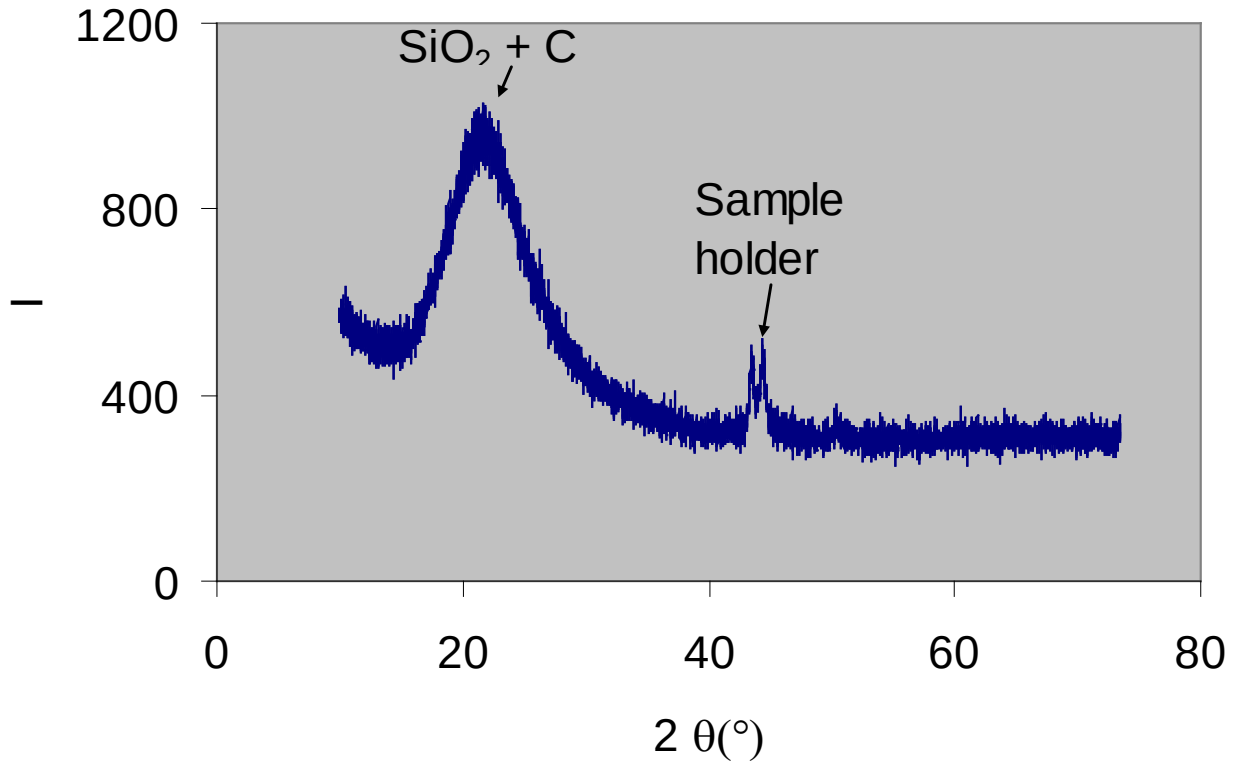
- **Parameters of the coating**
  - ✓**Thickness decrease**
  - ✓**Porosity volume increase**

Track formation

Vycor glasses + C

- Ar – 245MeV $\Phi=10^{13}$ cm^{-2}

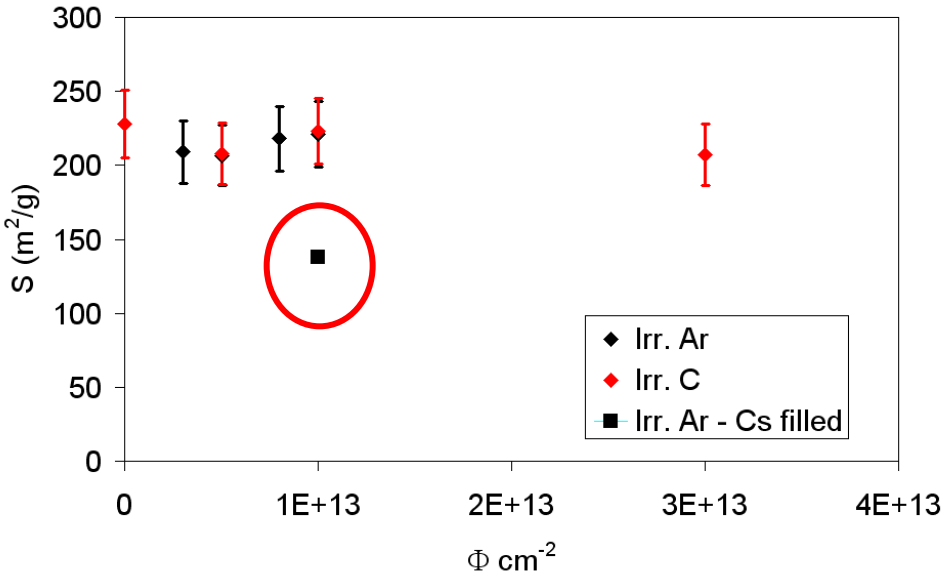


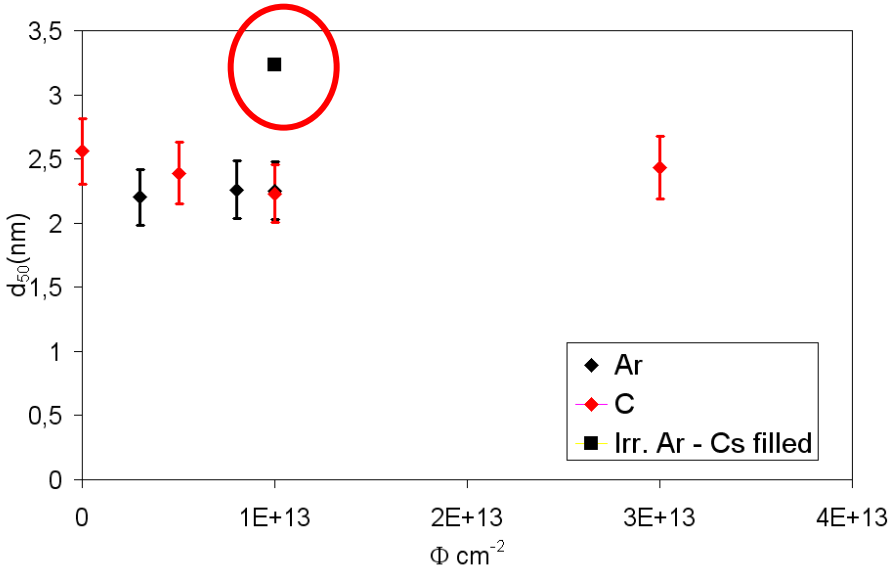


No reaction between silica and carbon

Vycor glass + Cs

Ar – 245MeV $\Phi=10^{13}$ cm^{-2}





- Lower specific area
- Higher pore diameter

→ Consequence of the Cs incorporation (chemical effects)

Conclusion

- Vycor glasses under irradiation
  - Reduction in the surface area in all cases
  - More ambiguous results for the pore diameter
    - Small increase with D and He
    - Small decrease with C, Ar

→ Greater effects are expected for higher fluences

- Mesoporous gel layer under irradiation
  - Destructuration of the silica network under Xe 92MeV
  - Consequence of ballistic effects will be investigated in Jannus facility

Thanks

Clara Grygiel, Isabelle Monnet (Ganil)
Thierry Sauvage (CEMHTI)
Patrick Trocellier, Yves Serruys (JANNUS)

And you for your attention…

Website of our institute : www.icsm.fr