Heavy ion irradiations of UMo7/Al nuclear fuels

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Outline

1. Presentation of UMo/Al nuclear fuels
2. Heavy ion irradiation of UMo/Al nuclear fuels: state of the art
3. Some results
    1. Methodological work on UMo7/Al
    2. Selection of the most interesting nuclear fuels
4. Conclusions and outlook

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UMo/Al nuclear fuel development: presentation

International treaty for non-proliferation:
^{235}U enrichment reduction down to 20% for nuclear fuel materials.

Research reactors:
UAl_x/Al or U_3Si_2/Al nuclear fuels enriched up to 93% in ^{235}U

For keeping high performances without designing new cores ⇔ new fuels have to be designed
^{235}U enrichment reduction must be compensated by an increase of the U density inside the fuel.

Density in U :
UAl_x : 4.3 g/cm^3
U_3Si_2 : 11g/cm^3
UMo7 : 15g/cm^3

Choice: UMo/Al metallic nuclear fuels

Research reactors

- **Material Testing Reactors**

OSIRIS-Saclay

- **Neutron sources**

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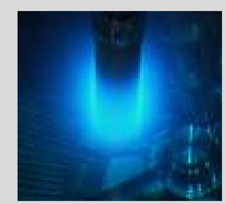
RJH-Cadarache

- **Neutron sources**



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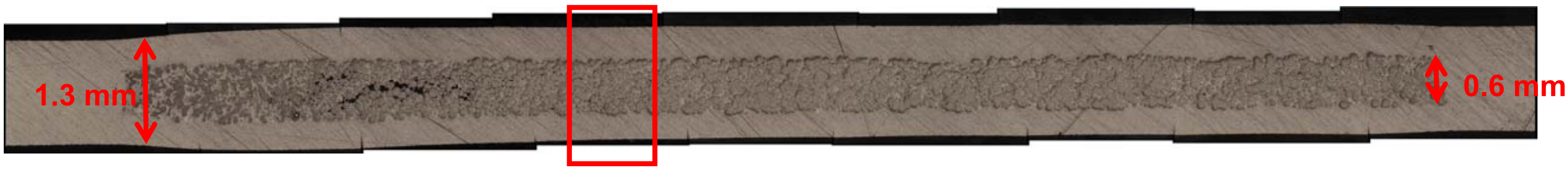
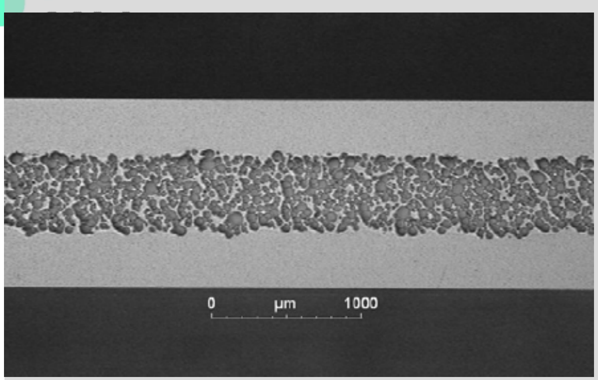
Research reactors

Research reactors in Europe (west) interested in UMo/Al for their conversion

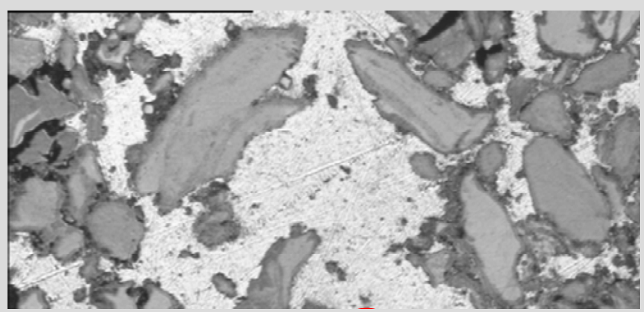
UMo/Al Nuclear fuel plate description

Cross sections

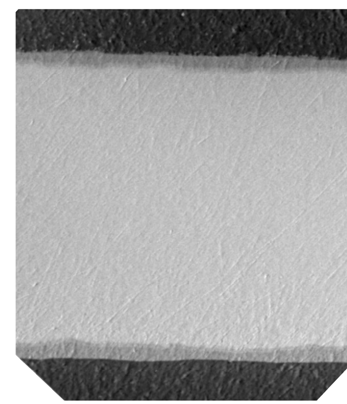




Atomized powder



Ground powder

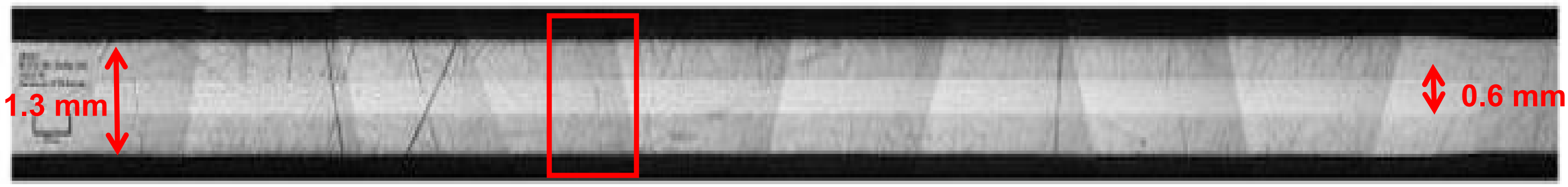


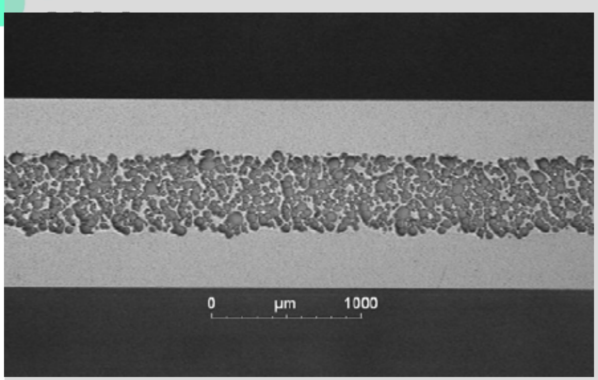
Monolithic

Two concepts depending on the required ^{235}U fission density (= ^{235}U density)

UMo/Al Nuclear fuel plate description

Cross sections





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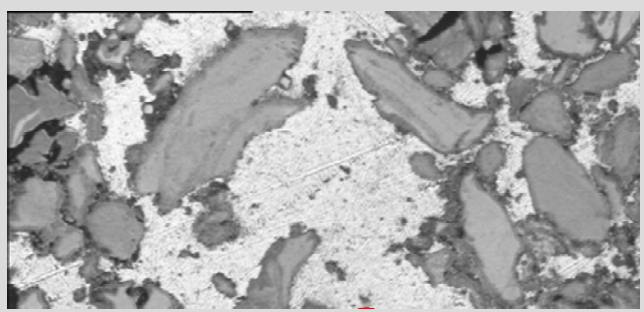

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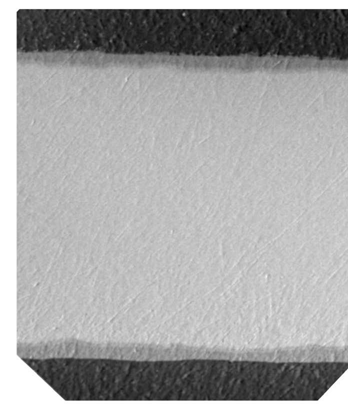

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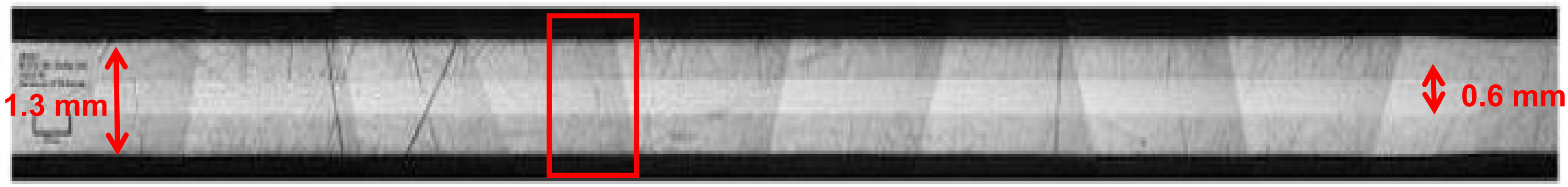

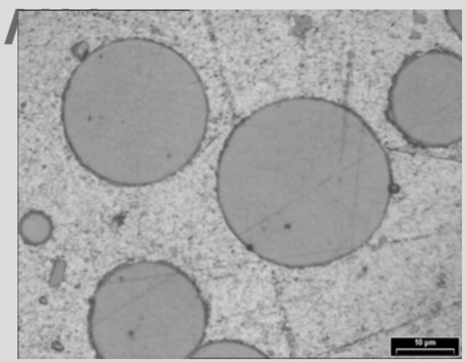

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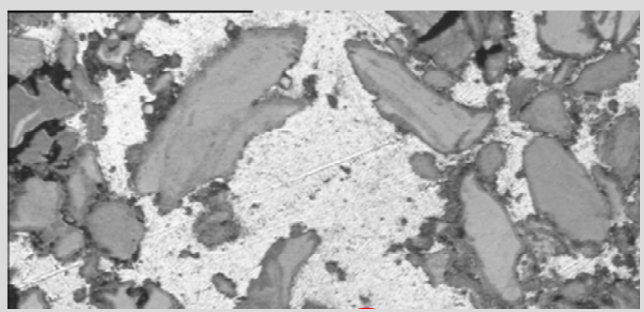

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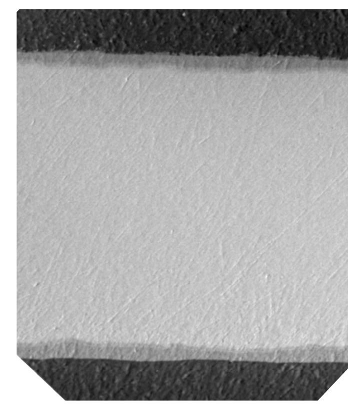
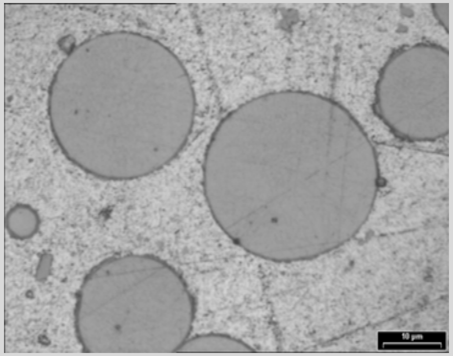


Monolithic

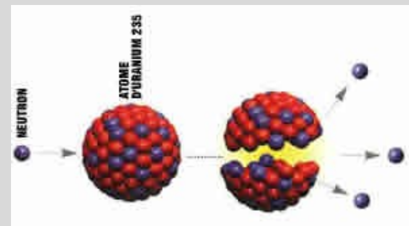
Two concepts depending on the required ^{235}U fission density (= ^{235}U density)

Nuclear fuel development: usual strategy

Design of new nuclear fuels



Test in dedicated nuclear reactors (Material Testing Reactors-MTR)

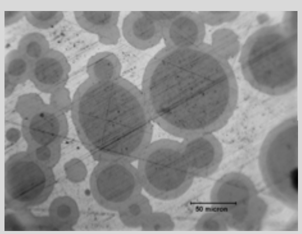


With increasing linear power

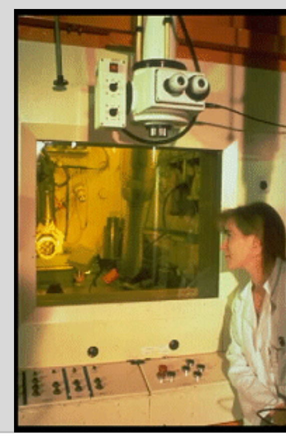
If not satisfactory

Post irradiation examinations (PIE) in hot labs (ex: LECA at Cadarache)

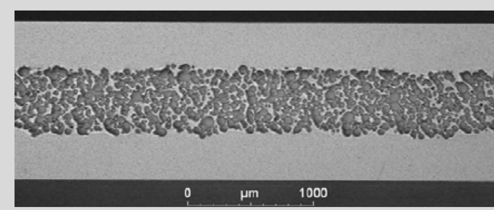
Destructive examinations



Microstructure evolution ….



Non- Destructive examinations

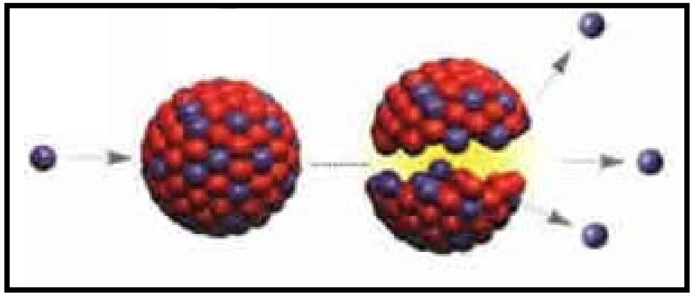


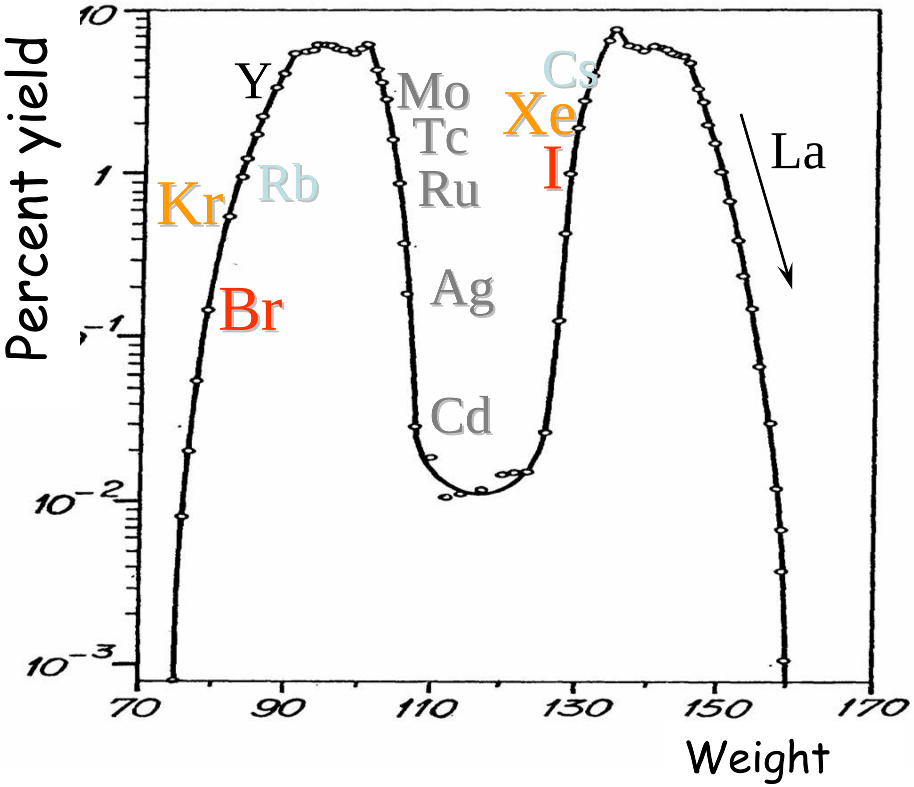
Swelling ?

Fuel plate behavior under in-pile irradiation

Irradiation = Fission

Radionuclides produced by ^{235}U fission with thermal neutrons





Fission products (FP):

• are emitted with a high energy; their energy loss in the fuel material will cause defects

Fission ⇔ defects' production

• are new radioelements (solid or gaseous) in the nuclear fuel material

Fission ⇔ production of new radioelements

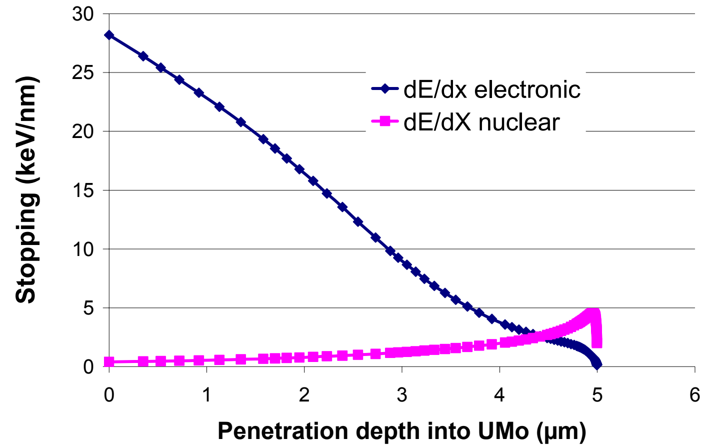
Fission ⇔ defects' production

^{235}U fission: about 200 MeV

(80% taken by the FP)

Penetration depth of Iodine (^{127}I of 80 MeV)

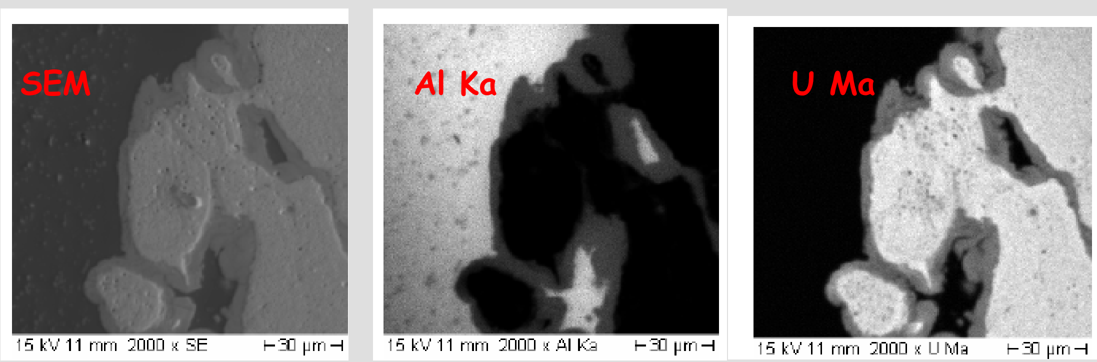
- in UMo: 5 μm,
- In Al: 15 μm.



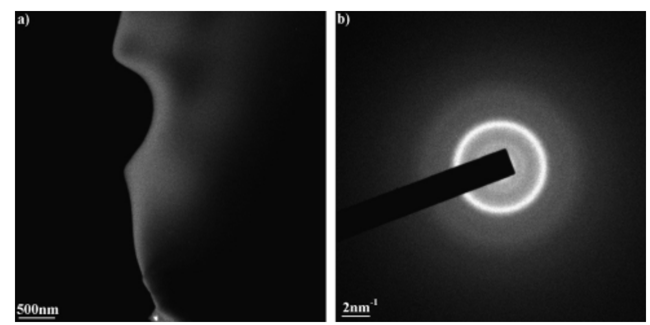
Irradiation temperature: often below 200°C

Irradiation induced diffusion:

- **Growth of an amorphous interaction layer at UMo/Al interfaces**



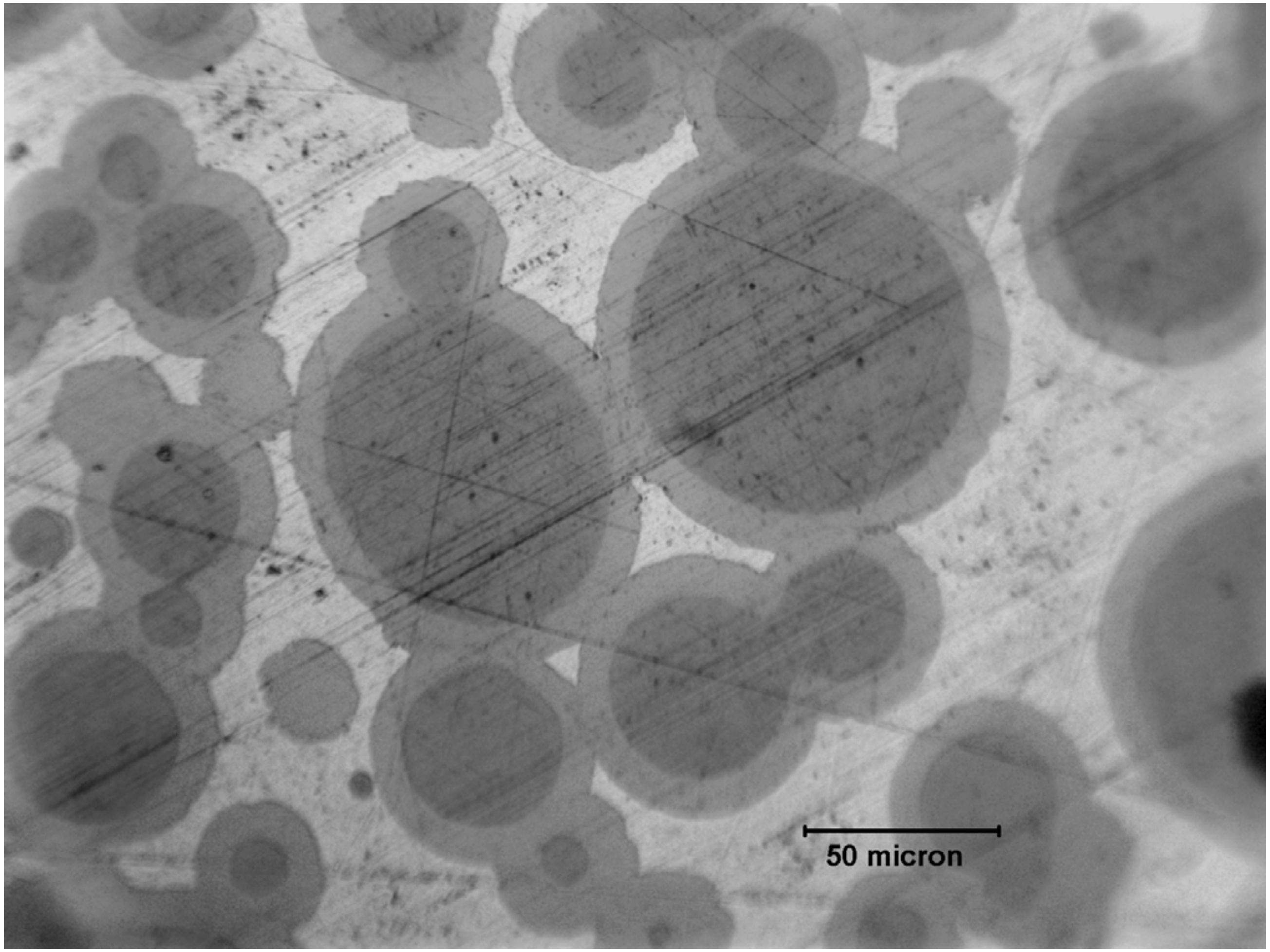
IRIS-TUM



FUTURE Irradiation

Van den Berghe *et al.*, *Journal of Nuclear Materials* (2008)

Fission ⇔ defects' production



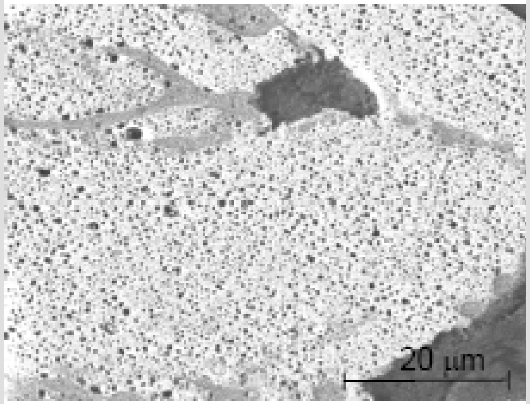
Micrograph taken from the RERTR 05 experiment

Fission ⇔ production of new radioelements

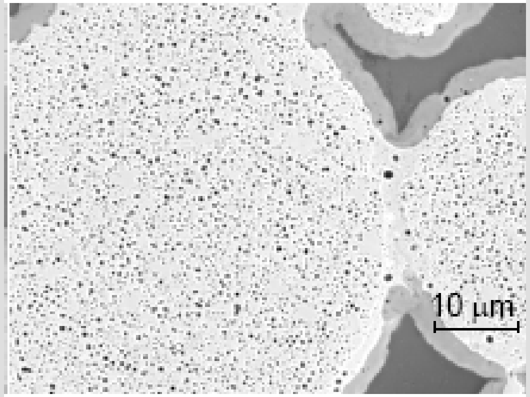
Gaseous fission products (Xe, Kr, …):

• Very low solubility limit into (formation of bubbles):

UMo

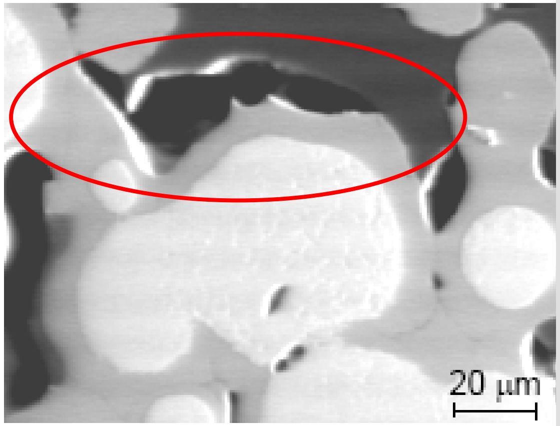


Ground powder



Atomized powder

UMo/Al interaction layer



• Good retention properties of the UMo phase for gaseous FP: low fuel swelling

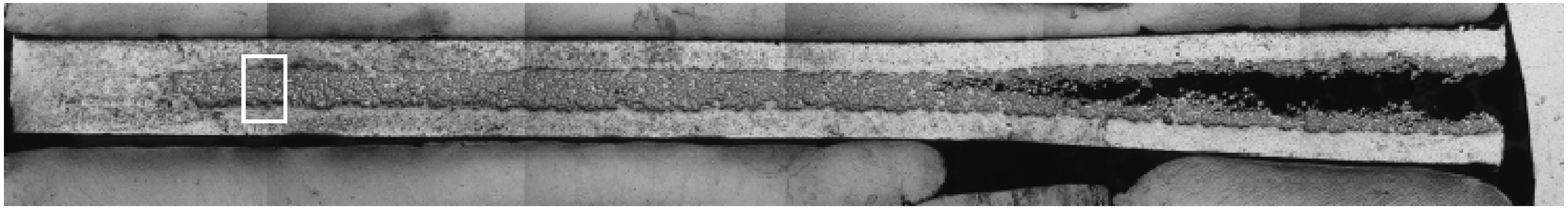
• Poor retention properties of the UMo/Al amorphous phase:

• Large porosities may interconnect and cause the breakaway of the fuel element (IRIS2)

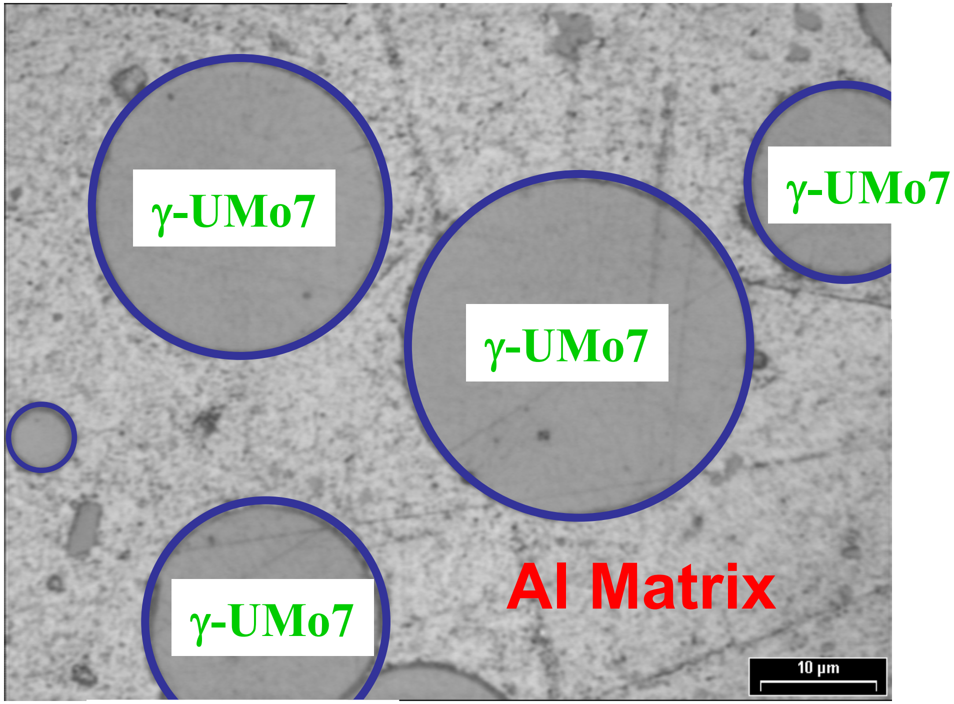
Fuel plate behaviour under in-pile irradiation

Cross section of a fresh fuel plate

Cross section of an irradiated fuel plate: breakaway



Technological solutions to test/improve

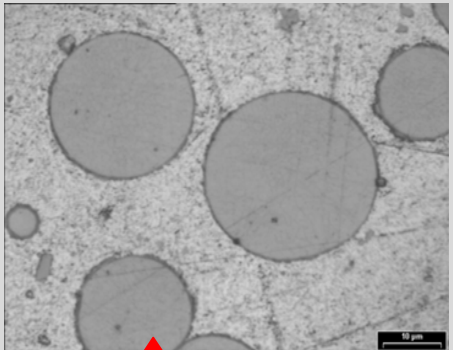


To limit the thickness and/or control the composition of the IL:

1. Matrix choice: limited adjunction of Si, Ti, …
2. Particles composition: UMo7, UMo10, adjunctions (Nb, Zr, …)
3. Anti-diffusion barrier: particle coating (Si, UO_2, …)

Nuclear fuel development: need for out-of-pile tests

Design of new nuclear fuels



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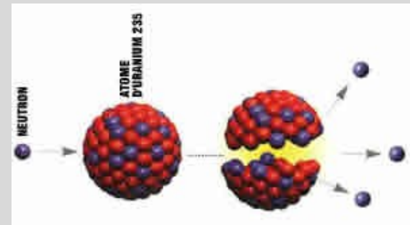

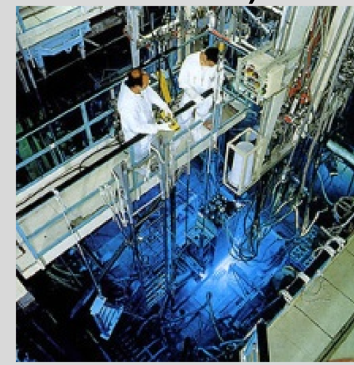

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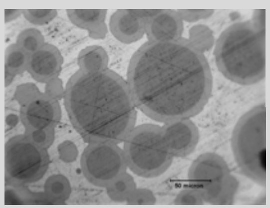

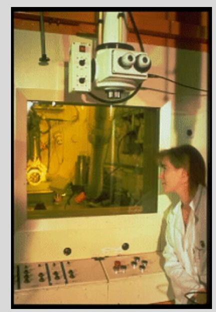

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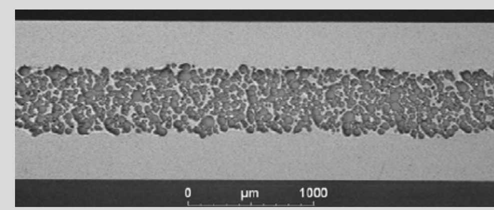

In-pile tests:

- Time consuming and expensive experiments,
- can be associated with out-of-pile activities

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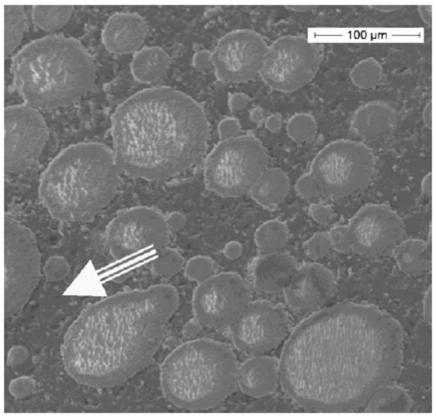
Simulation by out-of-pile methods

Demonstration of the interest of heavy ion irradiation for obtaining an UMo/Al interaction layer in 2005 (Wieschalla et al., JNM, 2006)



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Journal of Nuclear Materials 357 (2006) 191–197

journal of nuclear materials

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Heavy ion irradiation of U–Mo/Al dispersion fuel

N. Wieschalla [a,b,*], A. Bergmaier [c], P. Böni [b], K. Böning [a,b], G. Dollinger [c], R. Großmann [d], W. Petry [a], A. Röhrmoser [a], J. Schneider [e]

Heavy ion irradiation conditions:

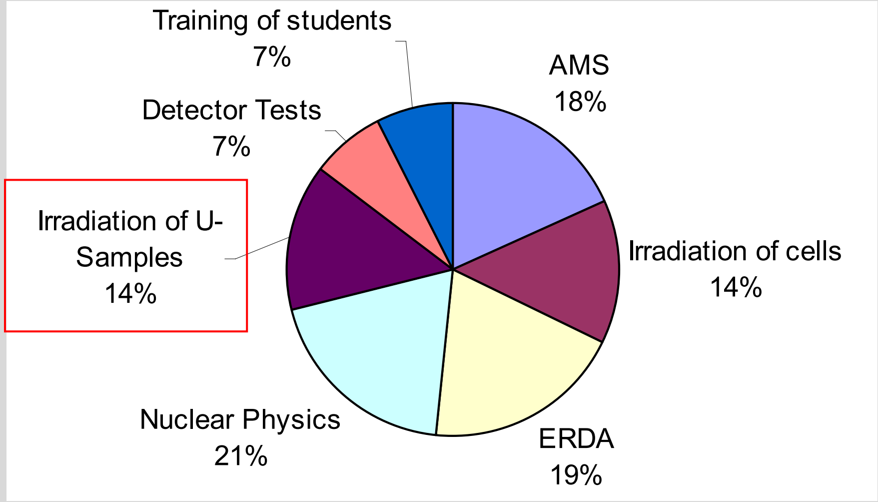
- projectile: ^{127}I with 80 MeV energy: typical ^{235}U fission product,
- irradiation angle: 30°

Simulation by out-of-pile methods

In 2005, start of a collaboration between the MLL, CEA and FRMII for:

- **methodological work on heavy ion irradiation for improving**
    - its reproducibility
    - our understanding of the influence of each experimental parameter (dose, flux, irradiation angle, …)
    - its “representativity” compared to in-pile neutron irradiation
- **technological solution discrimination studies**
  (H. Palancher et al., RRFM2006; R. Jungwirth (FRMII, Ph.D work))

Beam-time distribution at MLL in 2008



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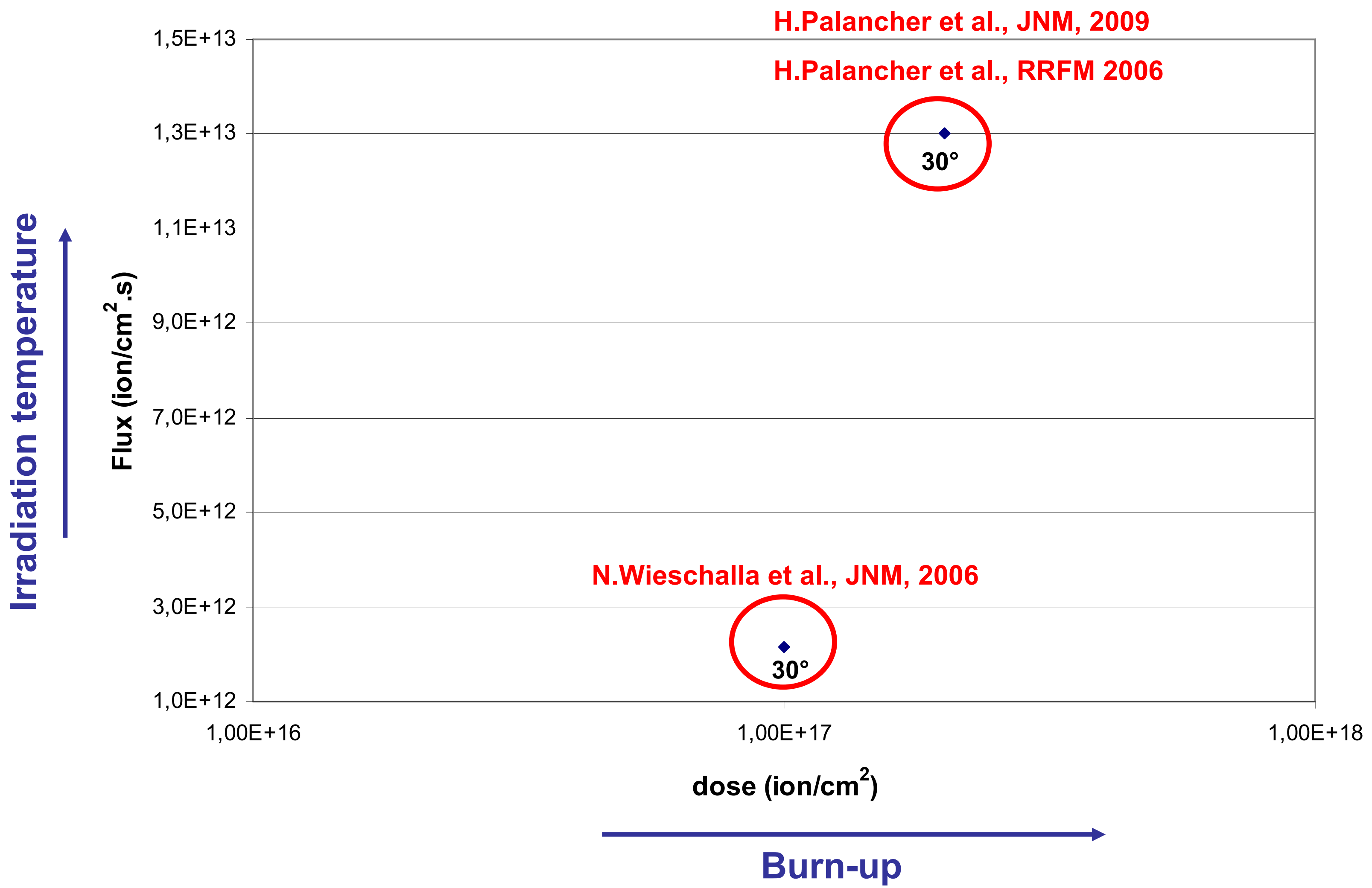
Heavy ion irradiation on UMo7/Al: 2008-2009 campaigns



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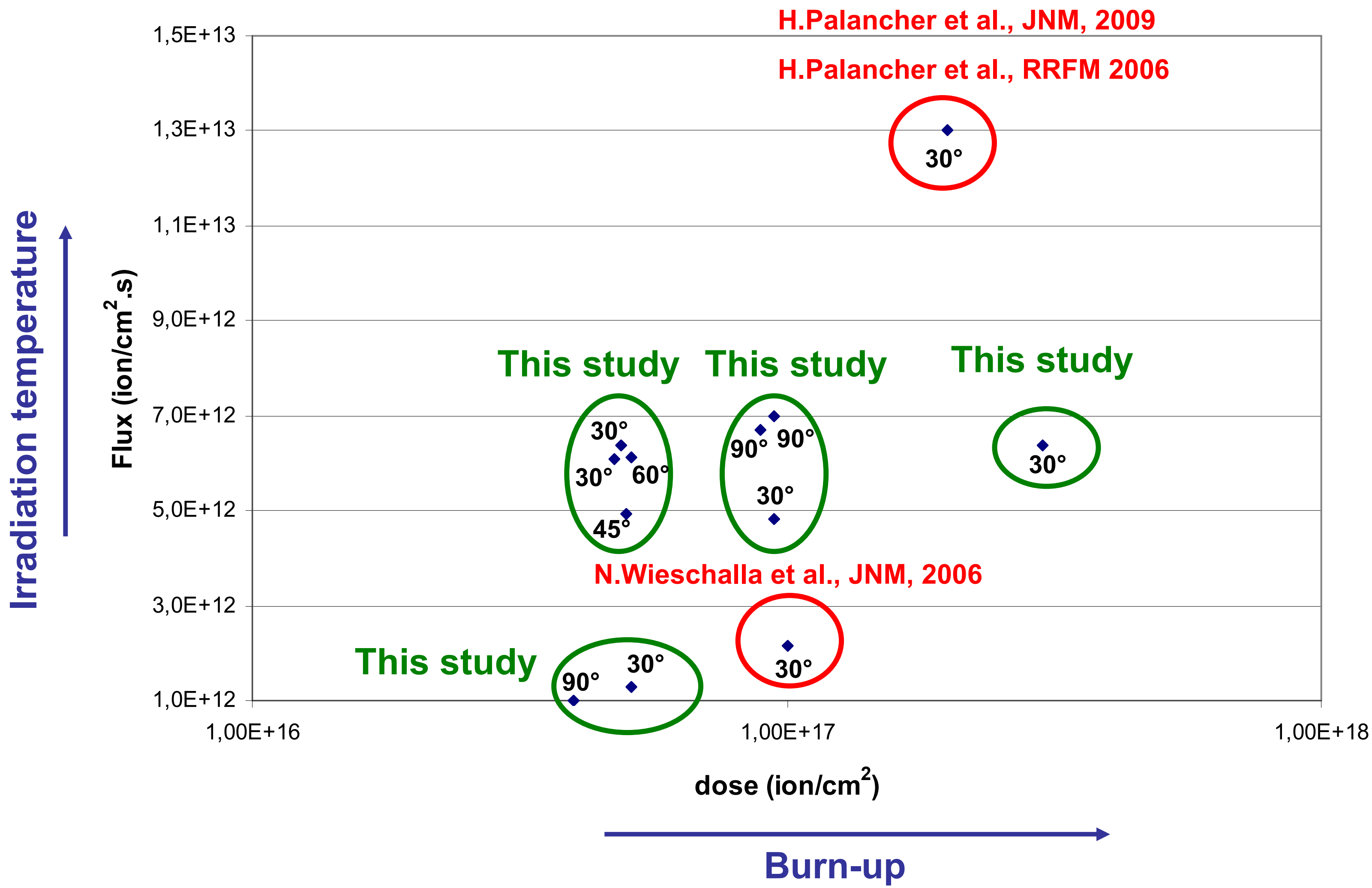

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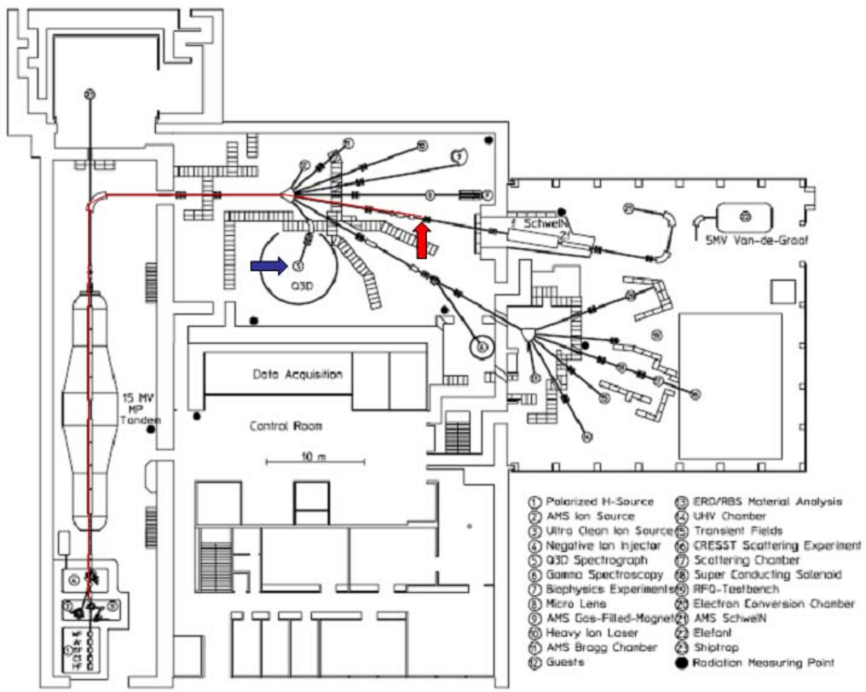
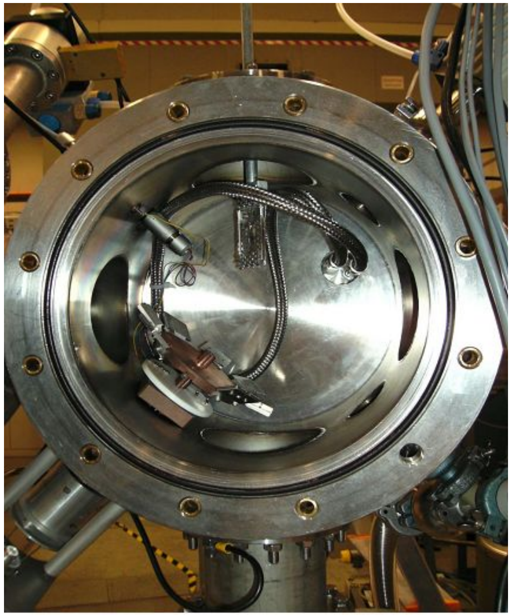
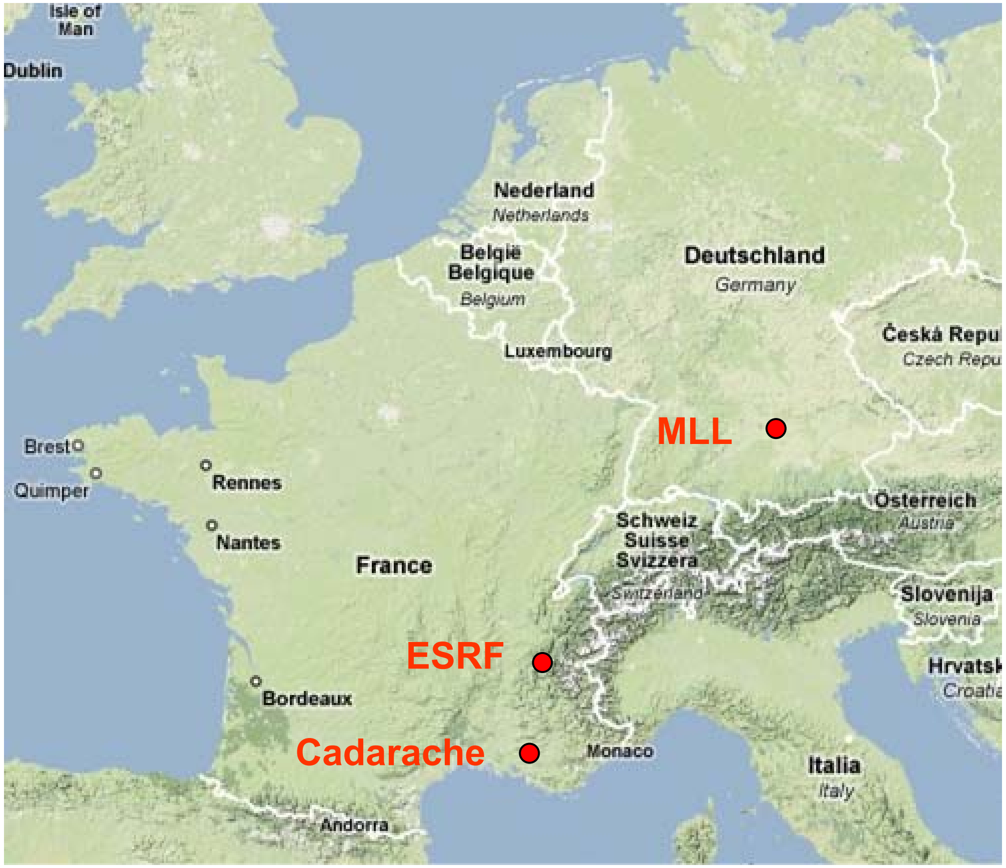




Many instrumental improvements have enabled:

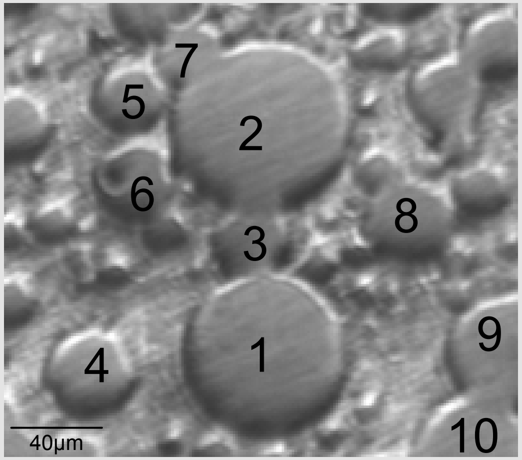
- Automation
- precise sample positioning in the beam,
- precise irradiation angle choice.

A European collaboration



Surface examinations

Before irradiation



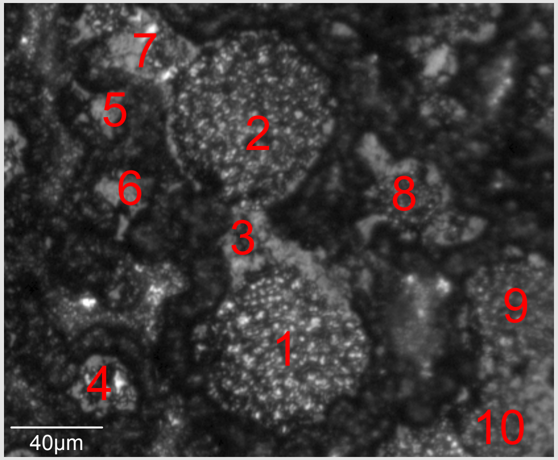
Optical

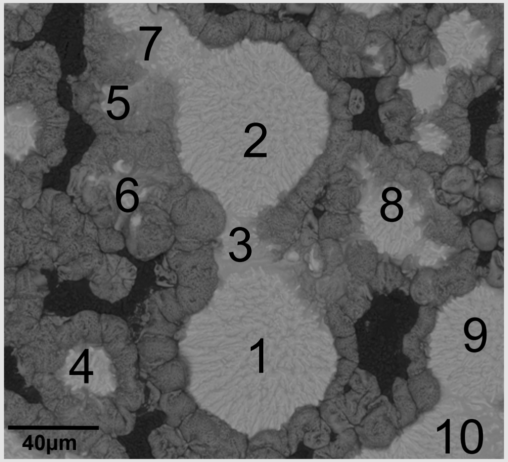
microscopy

After irradiation

Optical

microscopy





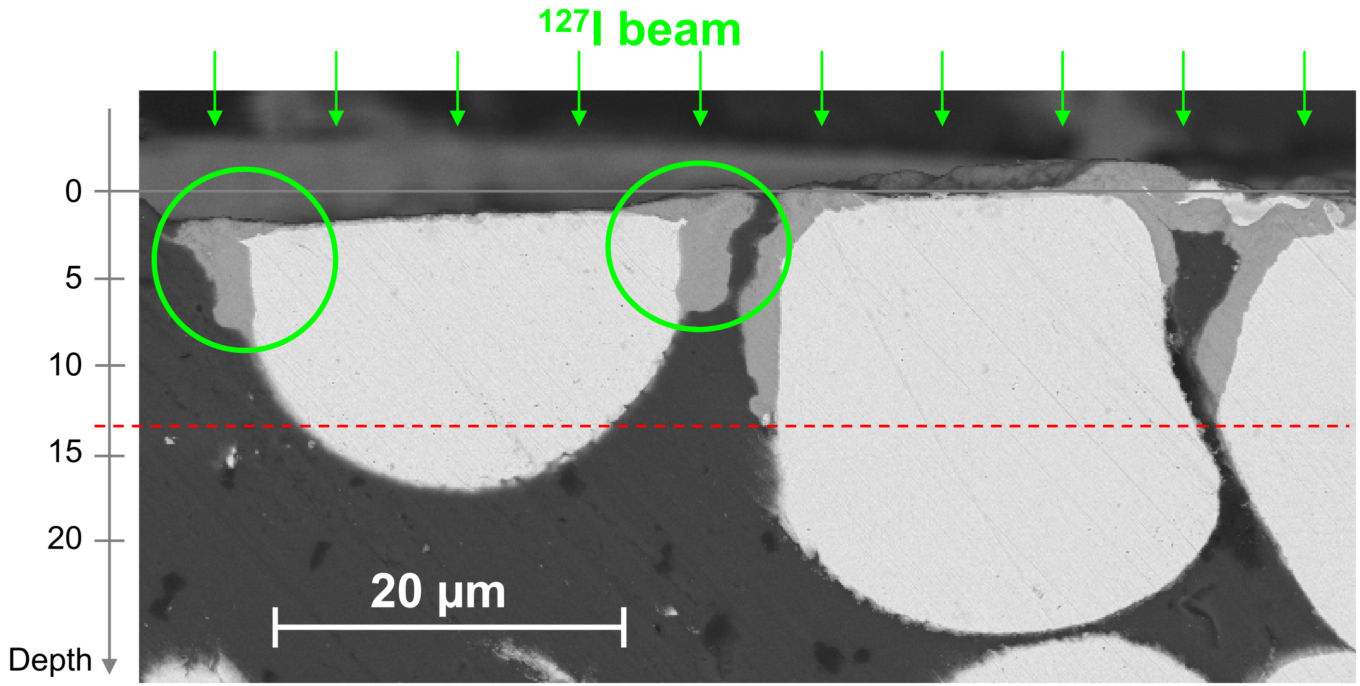
SEM

Whatever the irradiation conditions: an UMo/Al interaction layer has been observed

Transversal cross-sections examinations

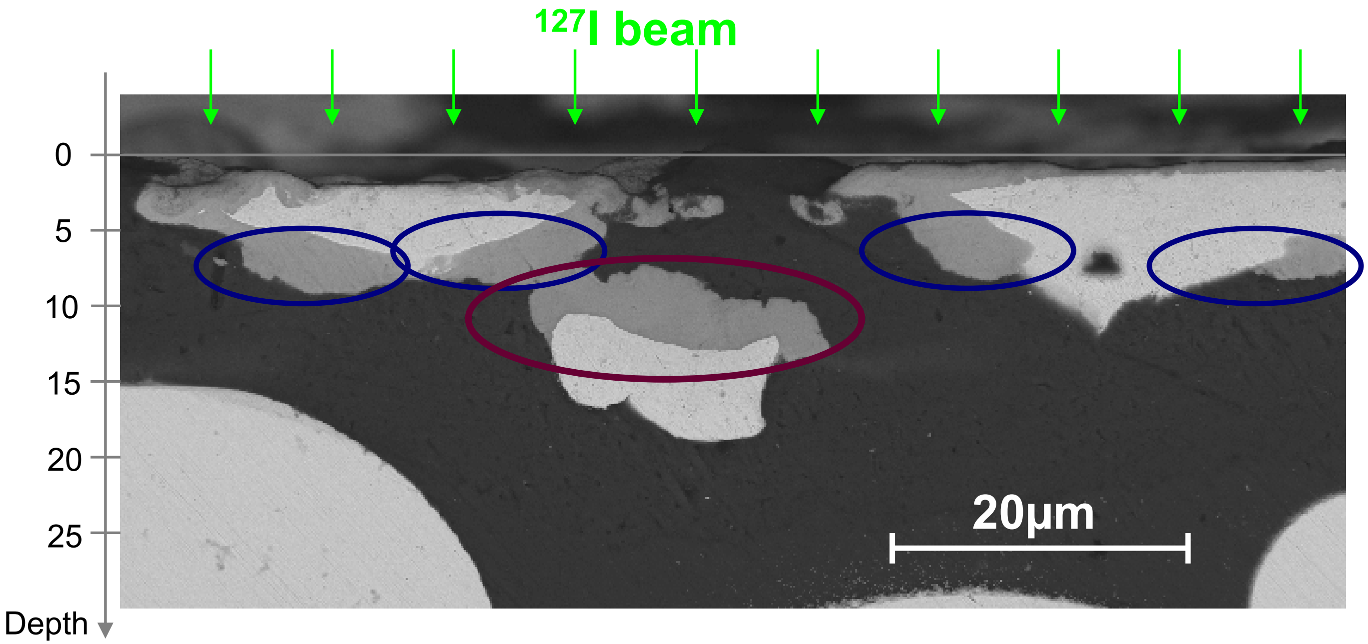
SRIM calculations

	^{127}I mean penetration depth (µm)
UMo	5,0 ± 0,7
Al	12,7 ± 0,6



The in-depth occurrence of the IL is in excellent agreement with the I penetration depth into the materials

Transversal cross-sections examinations

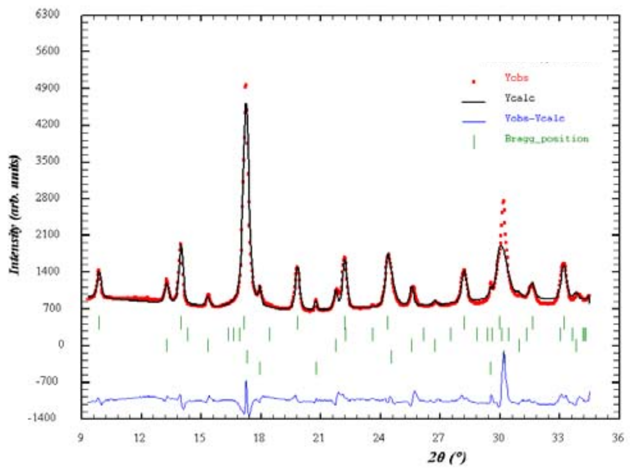


An IL has grown at each UMo/Al interface located at a depth compatible with I penetration depth. Both cases may be found:

-UMo/Al interface,

-Al/UMo interface.

μ-XRD studies of the IL composition



Example of μ-XRD diagram collected on the heavy ion irradiated zone of the fuel plates (data measured on the ID22 beamline)

	Weight fractions (%)				
	U(α)	UO_2	γ-UMo	Al	UAl_3
Irradiated area	0	7.1 (±0.3)	33.6 (±0.8)	22 (±1.0)	44.2 (±1.0)

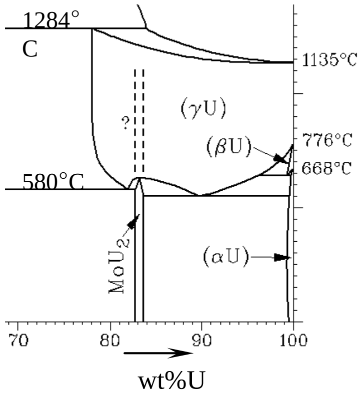
Conclusions of the μ-XRD study are consistent with previous studies (H.Palancher et al., JNM, 2009; RRFM2006):

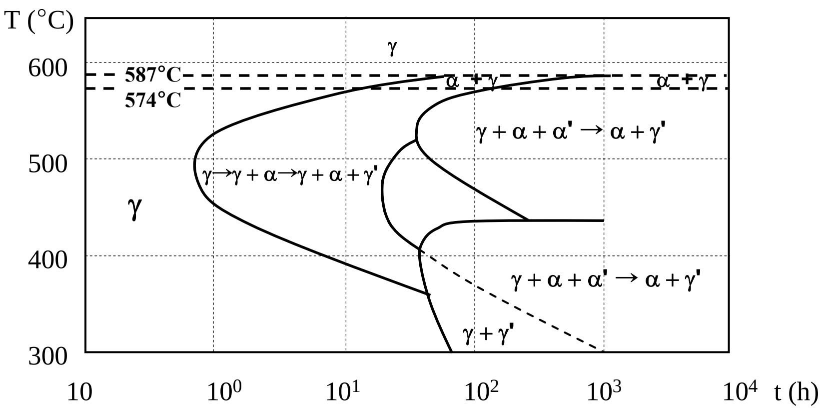
•The main component (UAl_3) of the IL is crystallised: its structure is however slightly modified/stressed (a cell parameter of about 4.23 Å is observed instead of 4.266Å)

•No information on the location of the Mo in the IL can be deduced.

UMo phases behavior under heavy ion irradiation

•Temperature influence on the stability of γ(U,Mo):





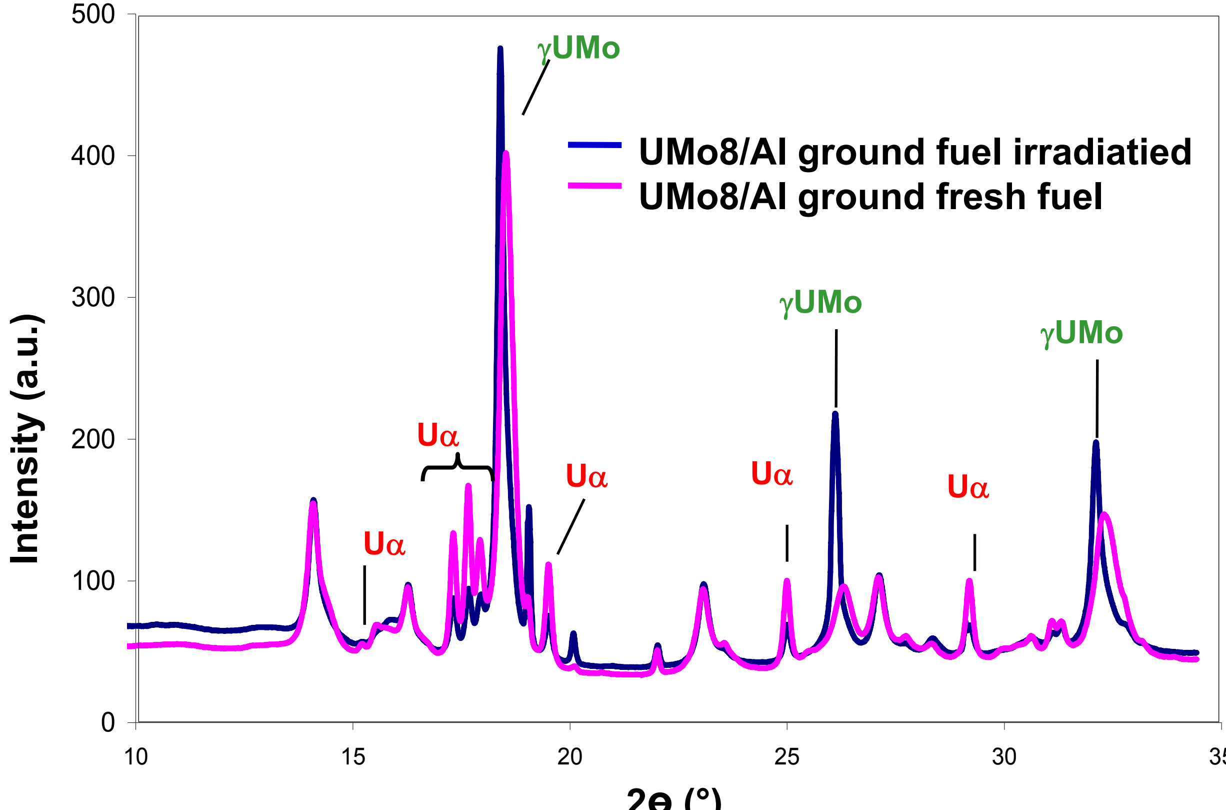
→ Destabilisation of the γ(U,Mo) phase for creating U(α) and U_2Mo or γ(U,Mo)

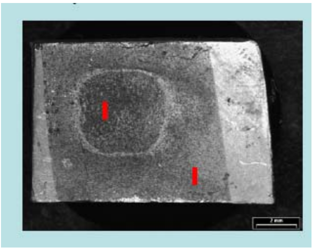
• Stability of γ(U,Mo) under in-pile irradiation at low temperature:

- XRD on IRIS1 fuel plate
- Studies from the 50's (see for example: M. L. Bleiberg, J. Nucl. Mater 2 (1959) 182 or S. T. Konobeevskii et al., J. Atomic Energy 1958 4- 1 p33-45)

→ **Stabilisation of the γ(U,Mo) phase: decrease of the U(α) weight fraction**

UMo phases behavior under heavy ion irradiation : UMo8/Al mini- plates





	Weight fractions (%)				U(α)/U(γ) (%)
	U(α)	UO_2	γUMo	Al	
Non irradiated area	20	27	43	10	47
Irradiated area	8	25	41	26	21

Stabilisation of the γ(U,Mo) phase: decrease of the U(α) weight fraction

Outline

1. Presentation of UMo/Al nuclear fuels
2. Heavy ion irradiation of UMo/Al nuclear fuels: state of the art
3. Some results
    1. Methodological work on UMo7/Al
    2. **Selection of the most interesting nuclear fuels**
4. Conclusions and outlook

Selection of the most interesting nuclear fuels

- **A set of 11 samples were irradiated in the same condition**

- **Most promising solutions:**
  - Si addition to the Al matrix confirmed by IRIS3, IRIS-TUM, RERTR06 in-pile irradiation.
  - UO_2 coating which currently tested in the IRIS4 experiment.

Dispersed fuel	Interaction layer characteristics	
	Presence	Thickness
UMo7 at / Al	YES	≈ 7 µm
UMo7 at / Al, Si	YES	heterogeneous
UMo10 at / Al	YES	≈ 5,7 µm
UMo10 at / Al, Si	NO	
UMo7 ox (µm)/Al	NO	
UMo7 ox (µm)/Al, Si	NO	
UMo7 ox (nm)/Al	Very limited interaction	
UMo7 ox (nm)/Al, Si	NO	
UMo7 /Al, Ti	Limited interaction	

Methodology : reference samples (green)

Oxide solution (grey)

Doped Al Matrices (magenta)

Conclusions

"Representativity" of Heavy ion irradiation versus in-pile neutron irradiation

- ILs obtained with heavy ion irradiation are not due to a temperature effect
- Heavy ion irradiations induce also the stabilization of the U(γ) phase as observed under low temperature in-pile irradiation,
- It has not been possible to obtain amorphous IL up to now: modifications of the set-up are undergoing.
- Doses must be increased to be more representative of the burn-up obtained in-pile

This method has been applied to a large extent to select best candidates to irradiate in-pile

Publications related to this work

H. Palancher, N. Wieschalla *et al*., J. of Nucl..Mater. 385, 449-455, 2009.
H. Palancher, N. Wieschalla *et al*., RRFM Sofia, 2006.
S. Dubois, H. Palancher et al., RERTR South-Africa, 2006
E. Welcomme, H. Palancher, R. Jungwirth *et al*., RRFM Vienna, 2009.

Thank you for your attention !