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Transmutation and irradiation on transmutation at HFR

Presented by:
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Contribution from:
NRG and CEA

Delft, 14th May 2019

SIXTH FRAMEWORK PROGRAMME

FI6W-CT-2004-516520
EUROTRANS

www.ec.europa.eu/jrc

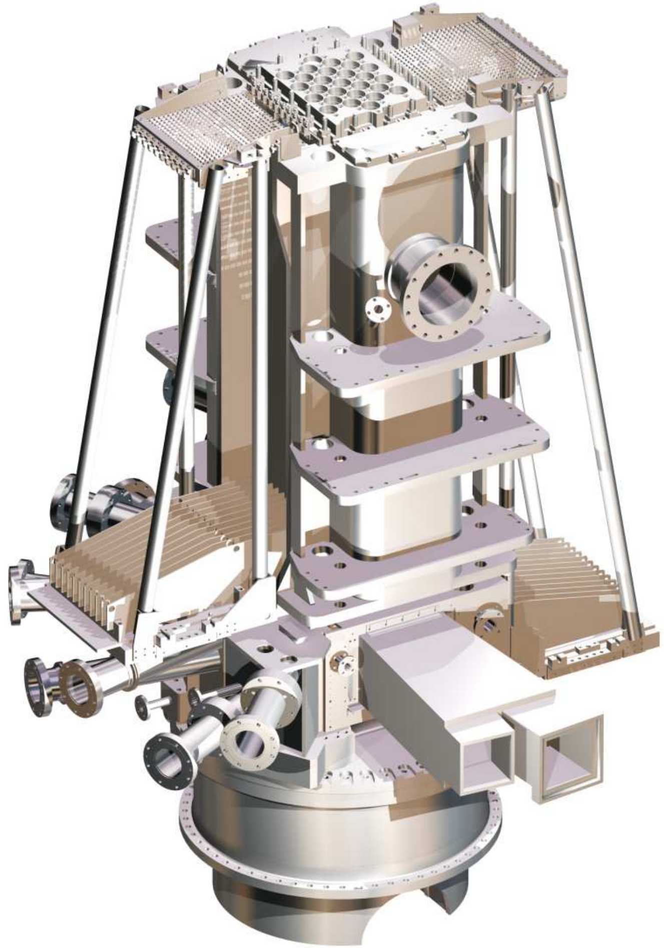
FairFuels
FAbrication, Irradiation and Reprocessing of FUELS and targets for transmutation

πelTriMM

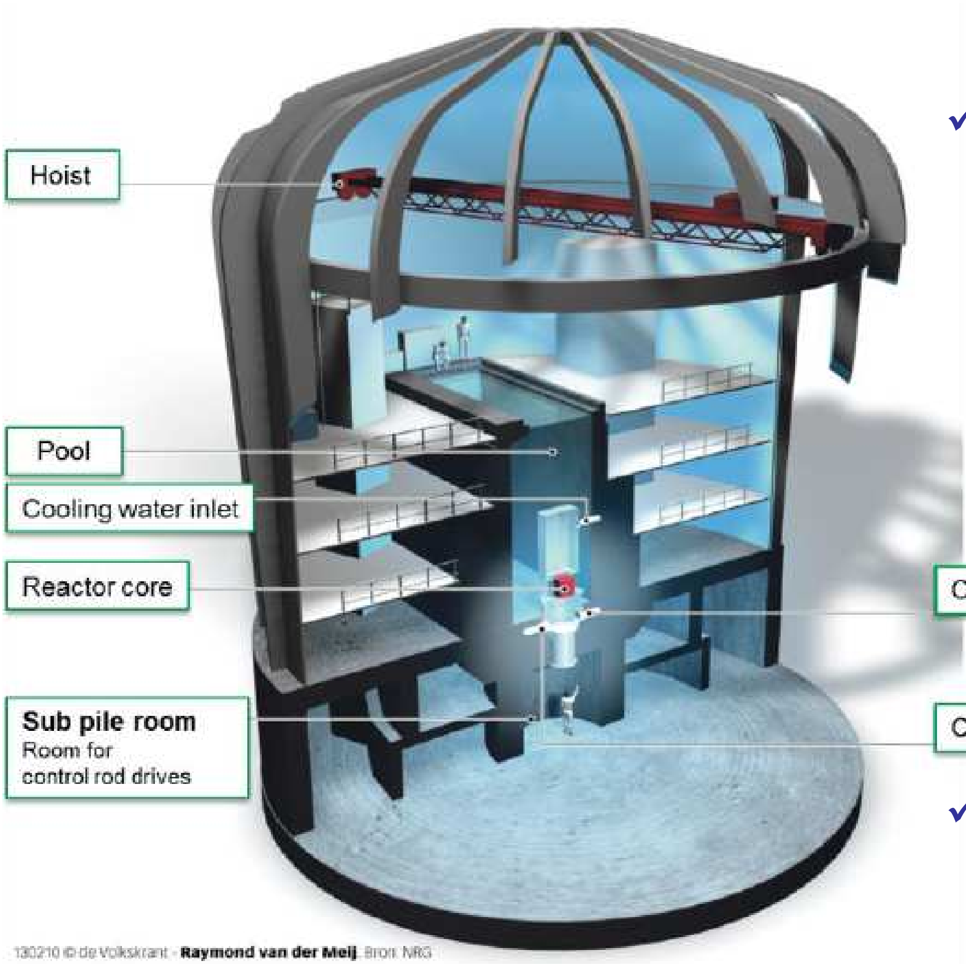
INSPYRE
Investigations Supporting MOX Fuel Licensing in ESNII Prototype Reactors

Synopsis

- HFR description
- Transmutation: Minor-Actinide irradiation tests.
- HELIOS, MARIOS, MARINE and SPHERE irradiations: purpose, description and status.
- Conclusions.



HFR General data



- ✓ High flux;
- ✓ 45 MW thermal power;
- ✓ Stable and constant flux profile in each irradiation position;
- ✓ Main applications:
  - Industrial and Medical radio isotope production
  - Nuclear energy irradiation services
  - R&D
- ✓ 31 operation days per irradiation cycle, 9 cycles a year

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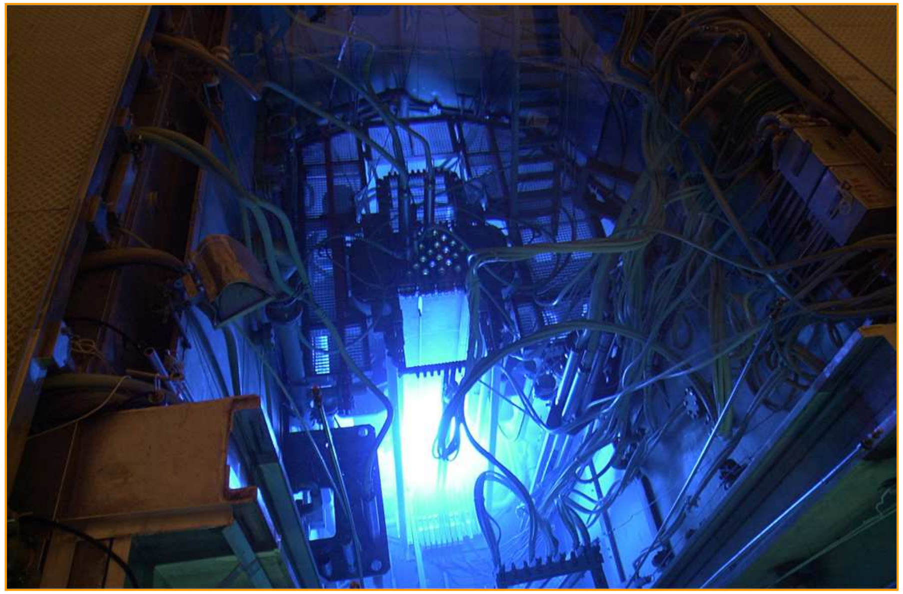
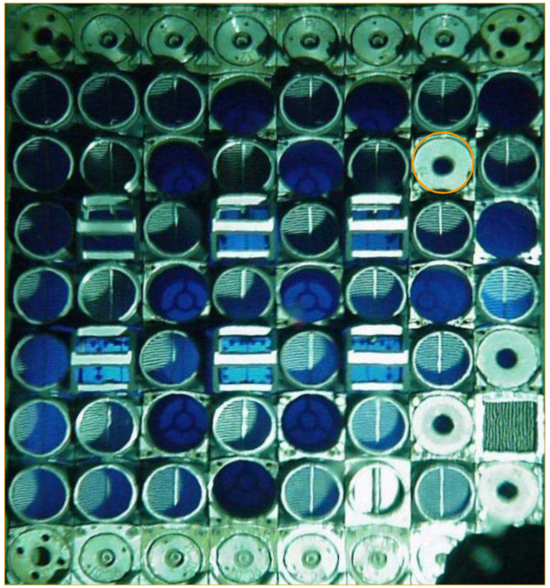
HFR General data

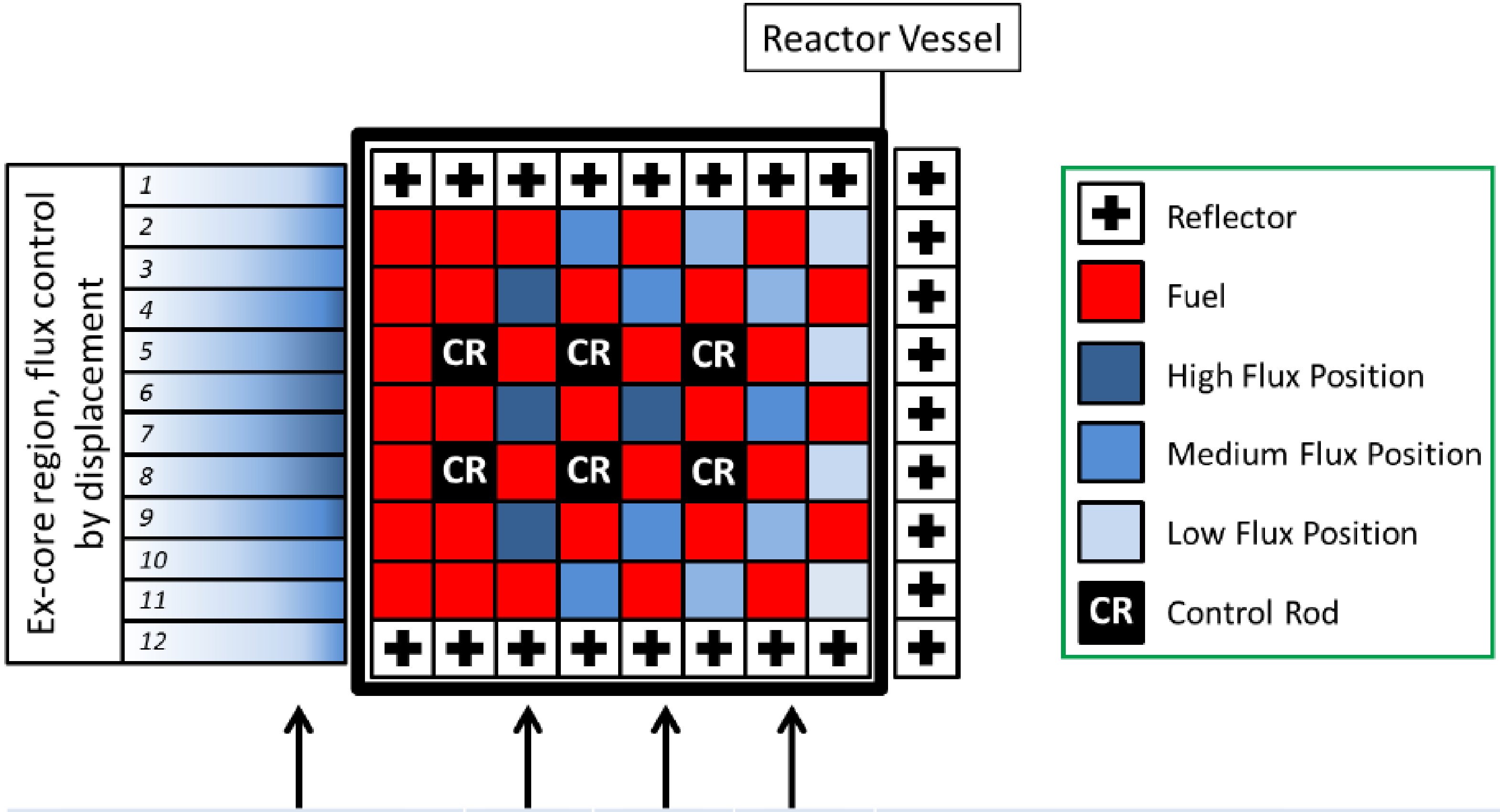


- Tank-in-pool multipurpose reactor
- First criticality in 1961
- Owner: European Commission
- Operator and License Holder: NRG
- License valid until at least 2025
- Availability approx. 290 days/year



- 9 × 9 core lattice
- 33 fuel assemblies (converted to LEU in 2006)
- 6 control elements
- 23 reflector elements
- 17 in-core positions + 22 PSF positions
- 12 neutron beams
- Useful height 600 mm
- Useful diameter 60 mm (65 mm in PSF)

HFR General data



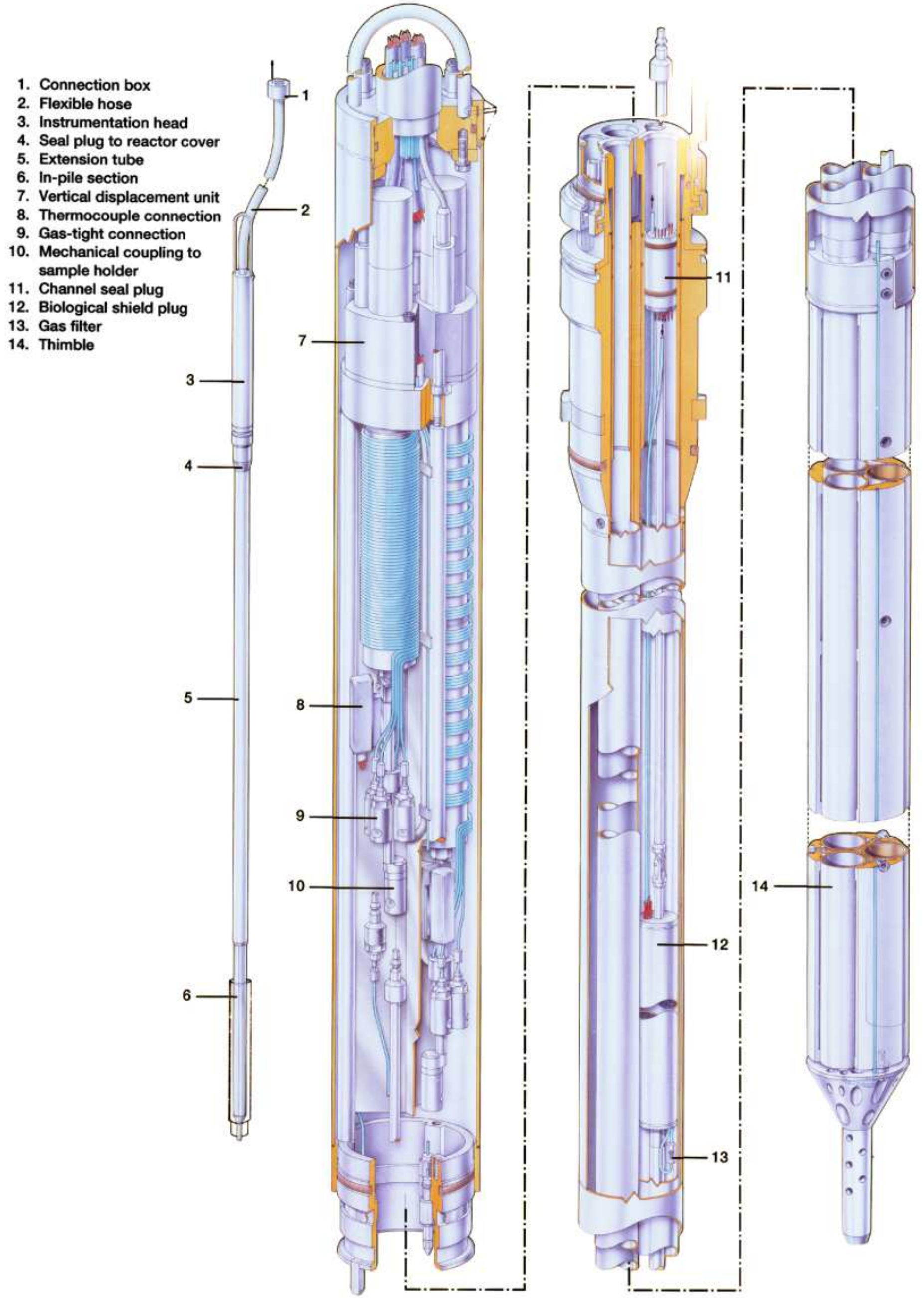
0-3, location dependent	5-8	3-5	1-3	Material DPA rate (DPA/year)
0-700, displacement controlled	n/a	500-700	300-400	Linear heat rate (W/cm fresh LWR fuel)

The stable and constant flux profile in each irradiation position is a unique HFR feature

HFR General data

Design and Modeling:

- Drawings
- Neutronics
- Thermal-mechanics
- Data analysis & interpretation
- QA/QC + X-ray lab
- Na handling lab
- Gas handling stations
- Multiple PIE options



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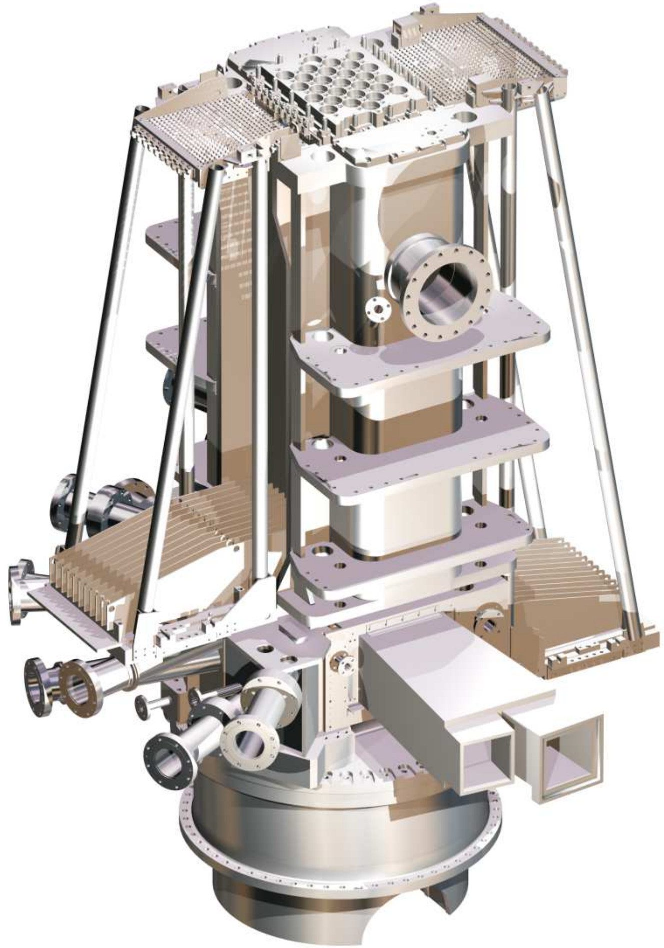

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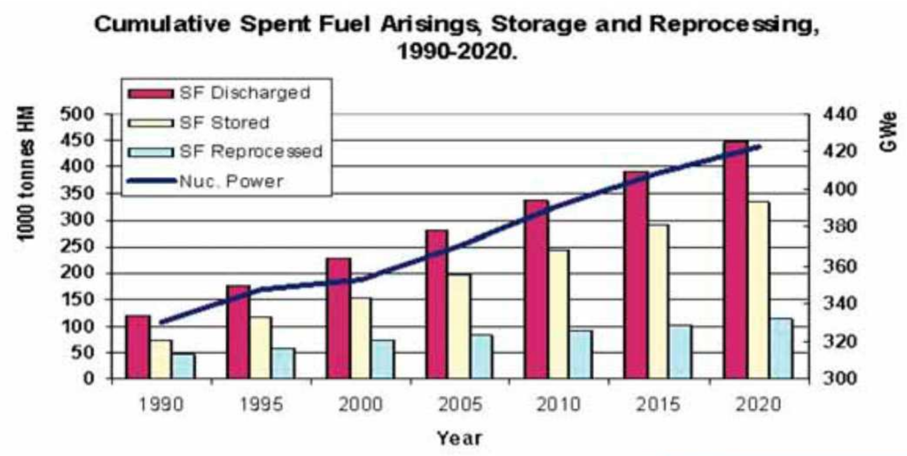
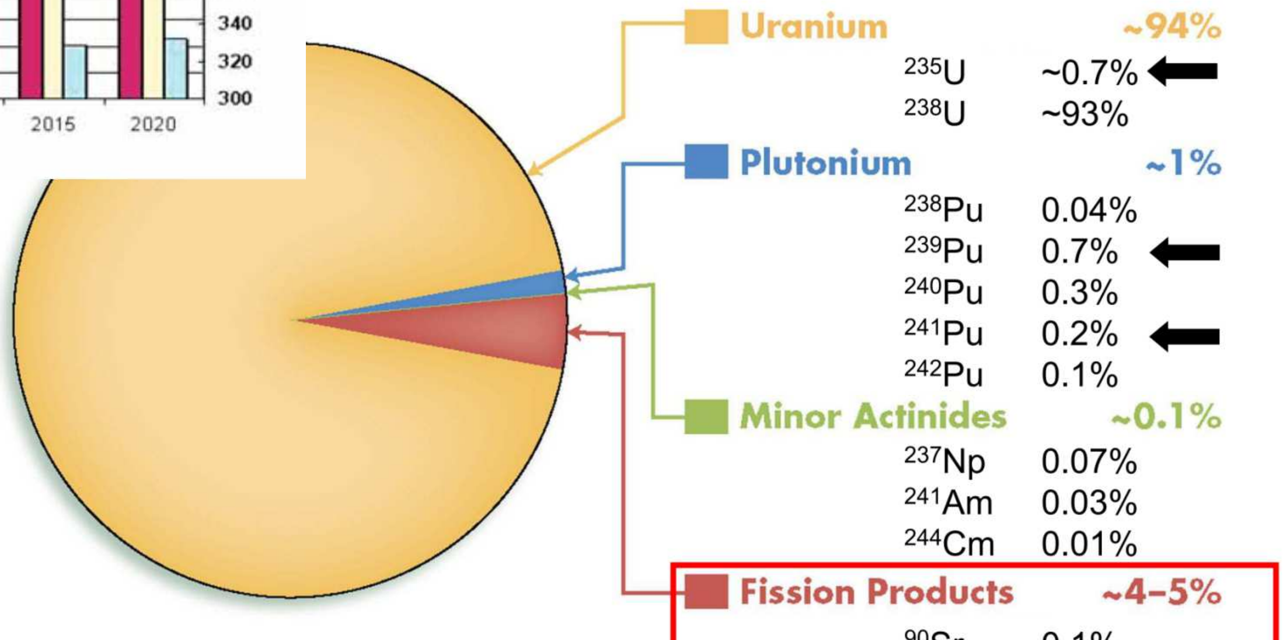
Transmutation: Minor Actinide Irradiation tests

- Spent fuel and energy demand are going to increase.



IAEA-TECDOC-1587 ISBN:978-92-0-103808-1

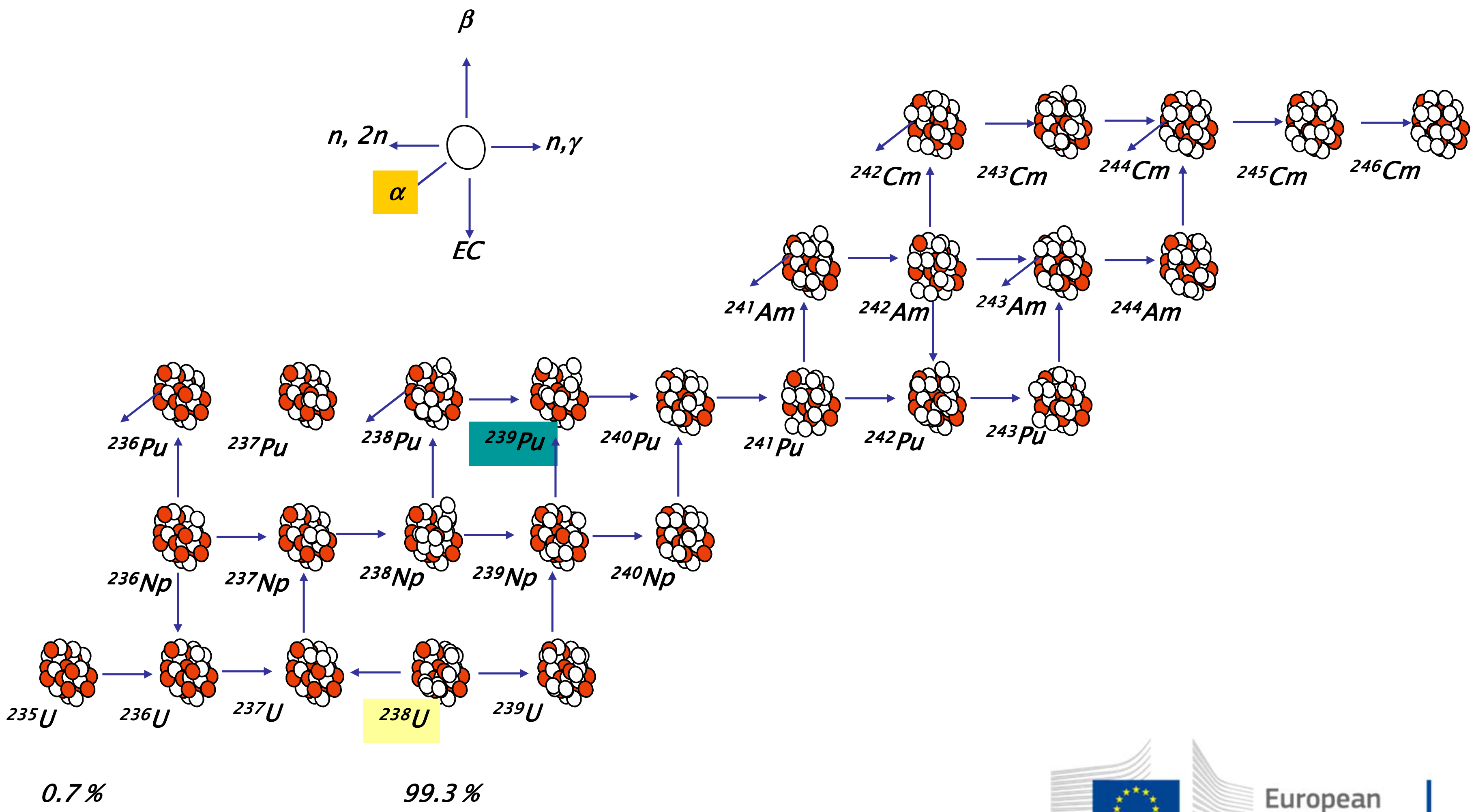


Approximate composition of used fuel

***From: Andrew Sowder, Advanced fuel cycle studies, presentation at IYNC 2012, Charlotte, USA**

Transmutation: Minor Actinide Irradiation tests

- Built-up of Minor Actinides by neutron capture.



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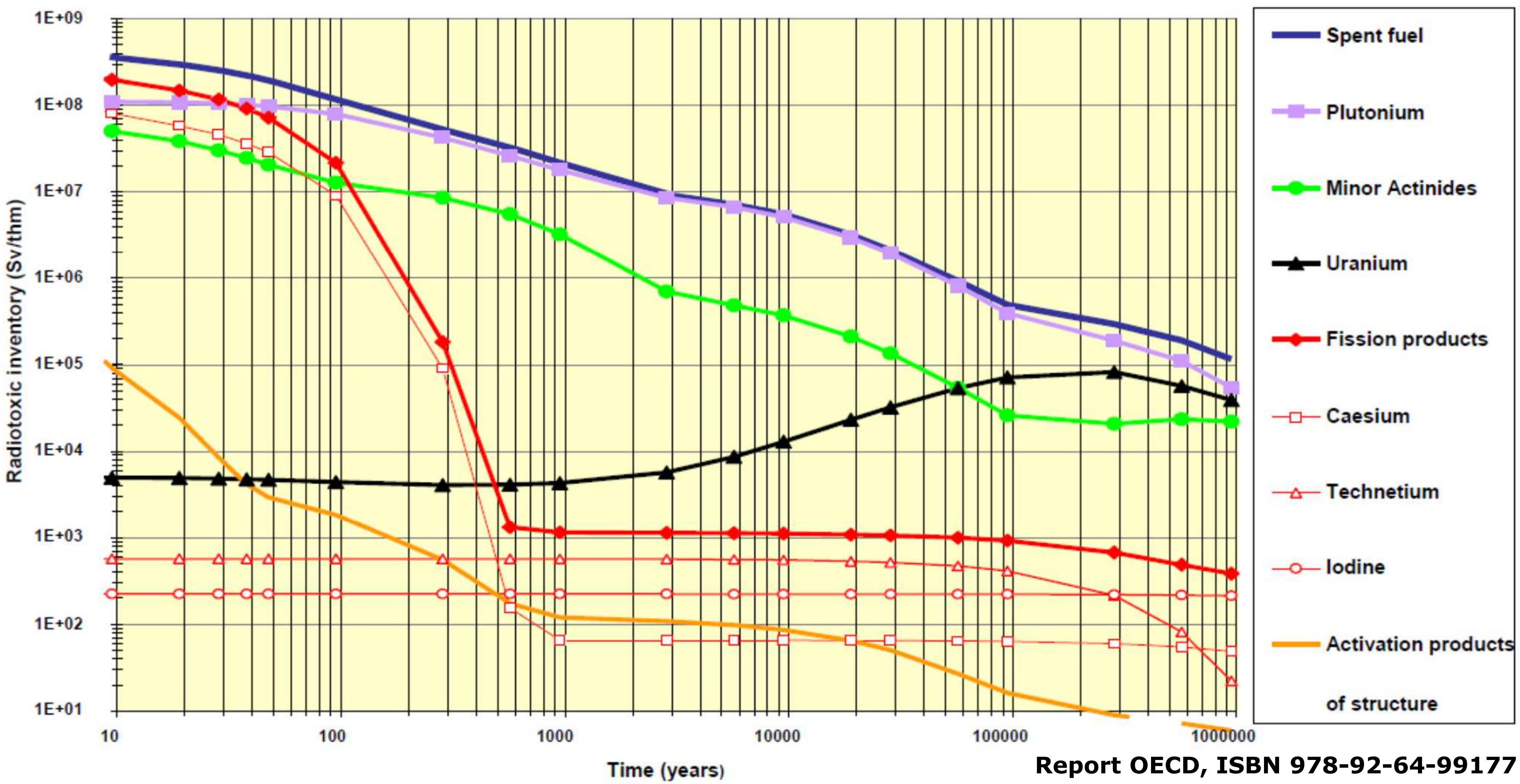
Transmutation: Minor Actinide Irradiation tests

- Transmutation of Minor Actinide (Am, Np, Cm) is going to be an option to reduce radio toxicity and footprint in the geological disposal for spent fuel.
- Otherwise, Finnish/Swedish concept foresees final disposal in granite.



Report OECD, ISBN 978-92-64-99177

Transmutation: Minor Actinide Irradiation tests

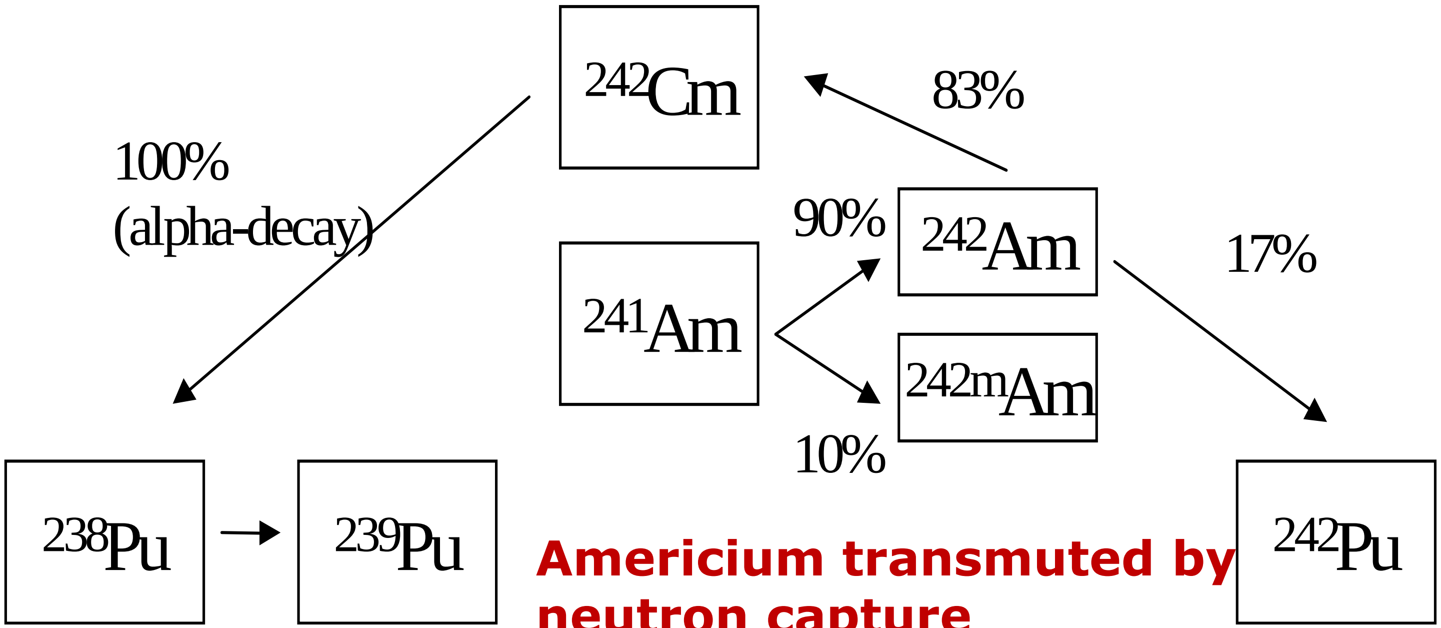
- Transmutation of minor actinides (MA) can be achieved using:
  - thermalised neutron facilities (LWRs)
  - fast neutron spectrum facilities,
    - critical reactors or
    - sub-critical accelerator driven systems (ADS)
- ADS operates in a flexible and safe manner even with a core loading containing a high amount of MA.

- Comparison of thermal and fast neutron spectrum.

	PWR UOX			FR (EFR)		
Isotope	σ_f	σ_c	$\alpha = \sigma_c / \sigma_f$	σ_f	σ_c	$\alpha = \sigma_c / \sigma_f$
^{237}Np	0.52	33	63	0.32	1.7	5.3
^{241}Am	1.1	110	100	0.27	2.0	7.4
^{243}Am	0.44	49	111	0.21	1.8	8.6
^{242}Cm	1.14	4.5	3.9	0.58	1.0	1.7
^{243}Cm	88	14	0.16	7.2	1.0	0.14
^{244}Cm	1.0	16	16	0.42	0.6	1.4
^{245}Cm	116	17	0.15	5.1	0.9	0.18
^{99}Tc	/	9	/	/	0.5	/

Transmutation: Minor Actinide Irradiation tests

- Americium is one of the radioactive elements that contributes to a large part of the radiotoxicity of spent fuels.
- Transmutation by irradiation in nuclear reactors of long-lived nuclides like ^{241}Am is therefore, an option for the reduction of the mass and radiotoxicity of nuclear waste.

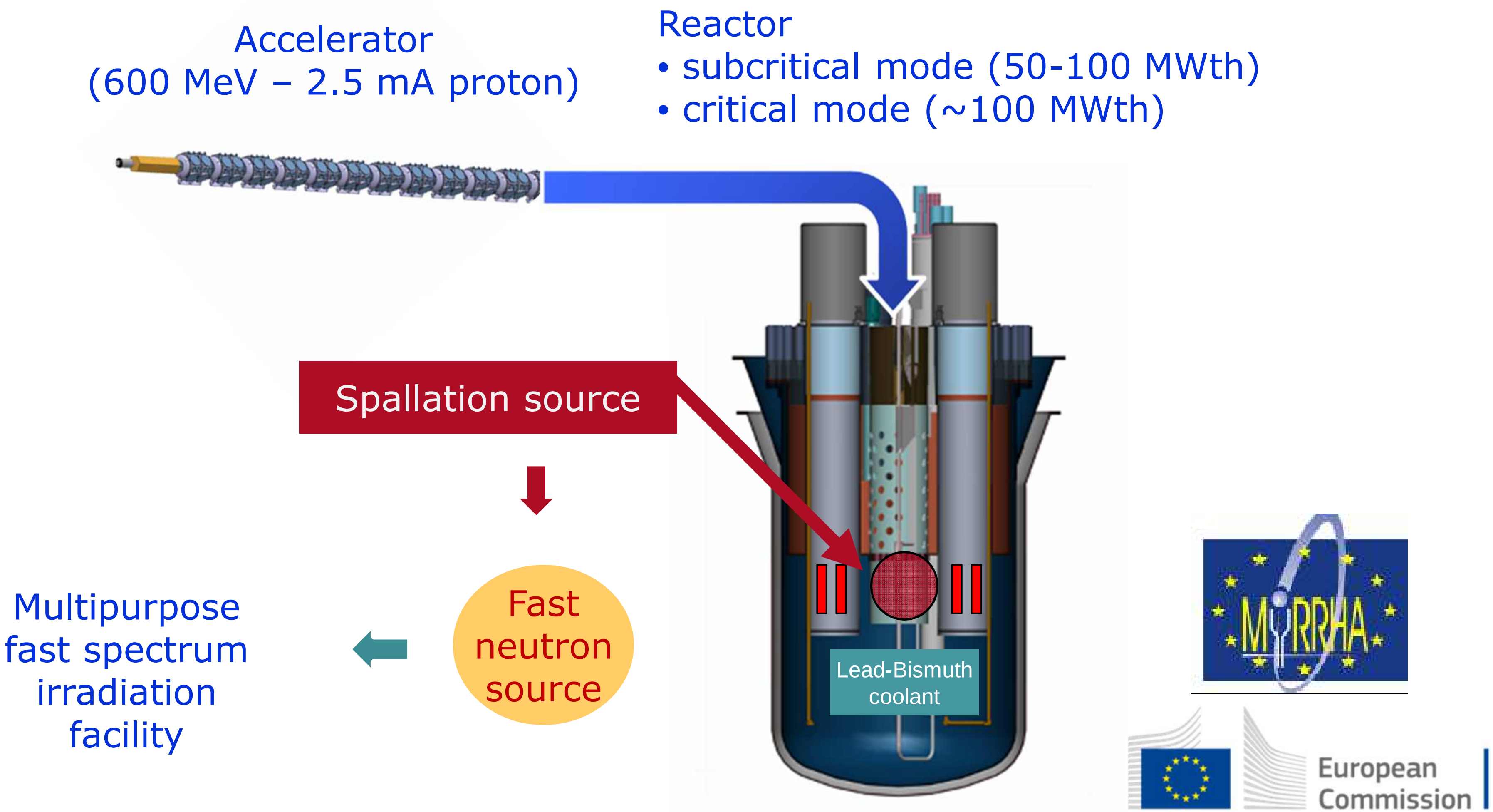


Americium transmuted by neutron capture

Transmutation: Minor Actinide Irradiation tests

ADS Transmutation principle:

- **HELIOS:** Test on U-free target containing Am. Cercer and Cermet concept



Transmutation: Minor Actinide Irradiation tests

Fast reactor strategy:

- Heterogeneous Recycling: Americium Bearing Blankets (AmBB)

  $(U,Am)O_{2-x}$ with ≈10-20% Am irradiated in the SFR radial blankets

  Demonstration of concept feasibility:

  - **MARIOS**: first separate-effect irradiation of AmBB in MTR

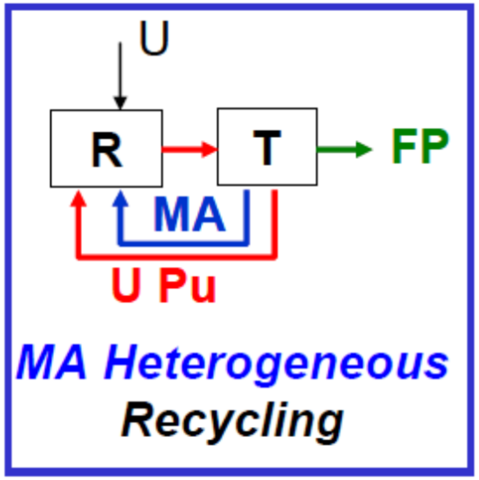
  - **MARINE**: first semi-integral irradiation of AmBB in MTR

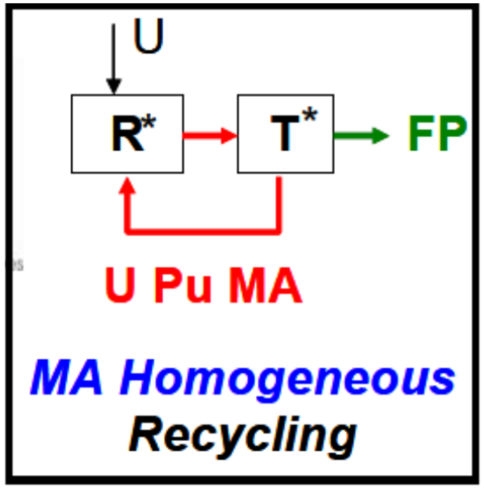
- Homogeneous Recycling: Minor Actinide Driver Fuel (MADF)

  A few percent of Am or MA diluted in the $(U,Pu)O_{2-x}$ driver fuel

  Prototypical qualification and optimization of the concept

- **SPHERE:** comparison of pelletized and sphere-packed fuel behavior → Optimization study (Spherepac)





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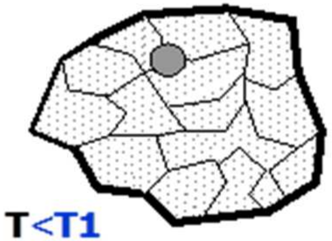
Transmutation: Minor Actinide Irradiation tests

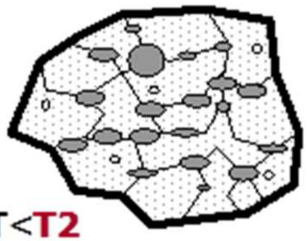


T<T1

500 to 700°C

He Implantation and annealing test

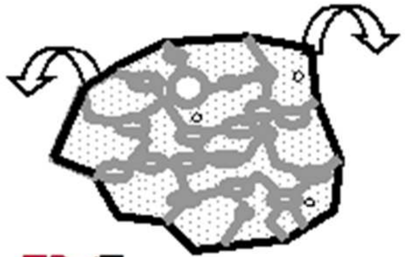
- ***Low He swelling (He atoms isolated in small size defects)***



T1<T<T2

- ***Potentially significant He swelling.***

MABB thermal conditions



T2<T

1100 to 1400°C

He Implantation and annealing
MOX annealing
SUPERFACT

- ***Low He swelling because of significant release***

0.75 He atom per Am-241 atom. 5x more gas production compared to LWR fuel

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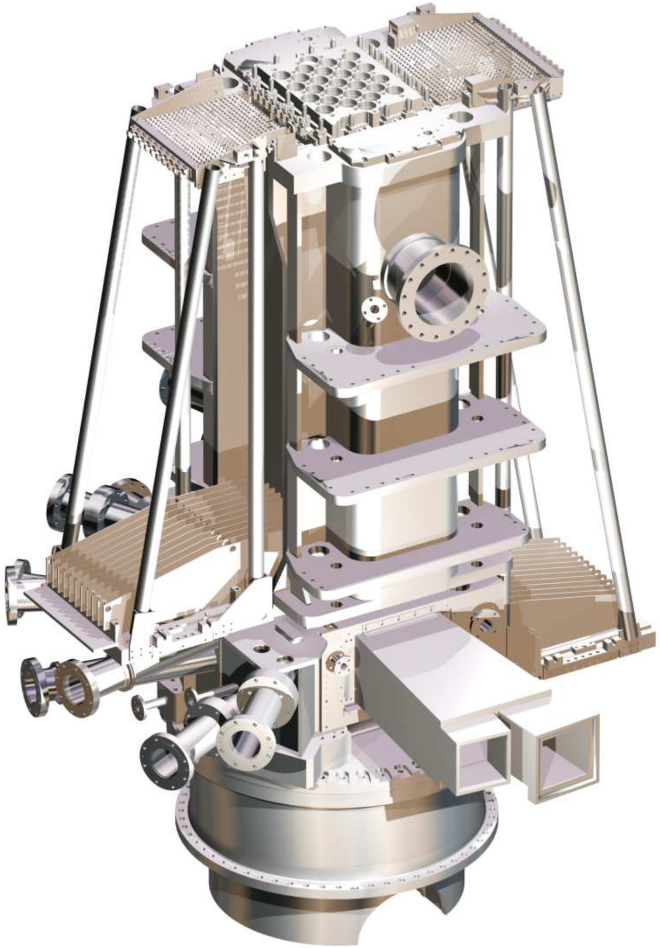

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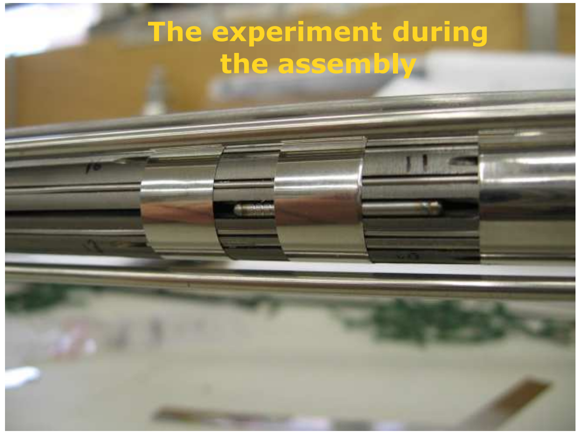
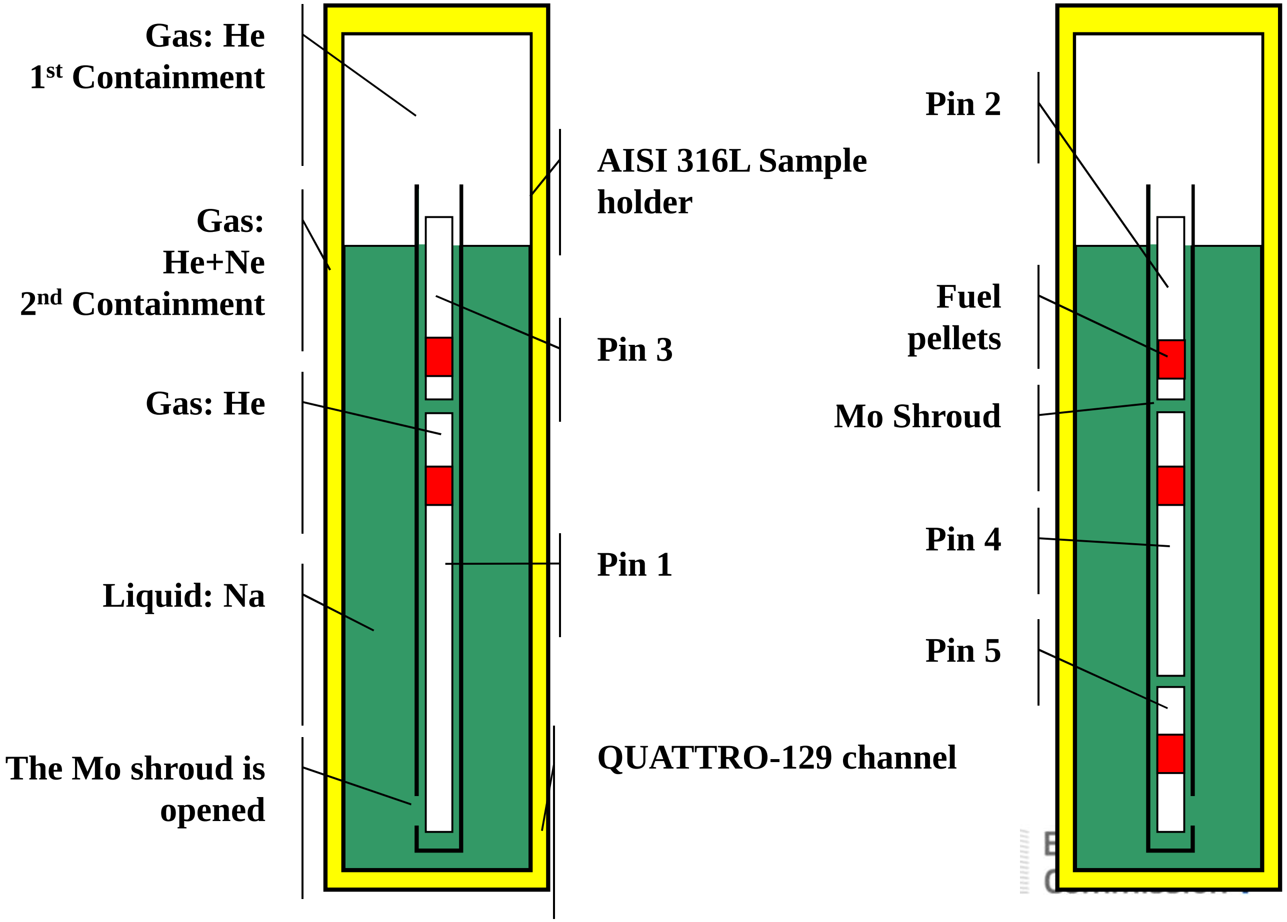
HELIOS irradiation test

Irradiation at HFR with CEA and NRG HELIOS (EUROTRANS, FAIRFUELS)



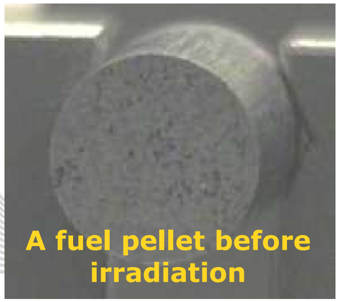
- **Objective:** Determine performance limits of MA incineration. Investigate temperature dependence of fuel swelling and gas release for uranium-free nuclear fuel containing Minor Actinides.
- **Status:** irradiated between April 2009 and February 2010. PIE completed.



HELIOS irradiation test

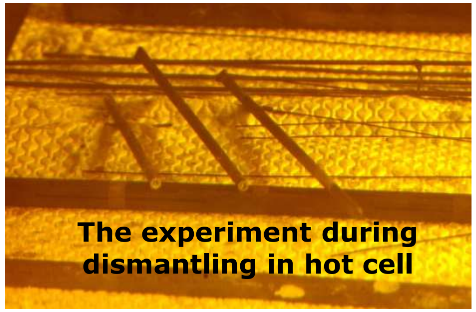
Pin Nr.	Composition	Microstructure	%Pellet TD Measured	As-fabricated density [g cm^{-3}]		Instr.
				^{241}Am	Pu-tot	
1	$Am_2Zr_2O_7$ + MgO	5-50 μm	91.5	0.66		
2	$(AmZr,Y)O_2$	Solid solution	92.6	0.7		TC
3	$(Am,Pu,Zr,Y)O_2$	Solid solution	89.7	0.74	0.39	TC
4	$(Zr,Am,Y)O_2$+Mo	65-125 μm	94.1	0.7		
5	$(Pu,Am)O_2$+Mo	>150 μm	96	0.3	1.2	

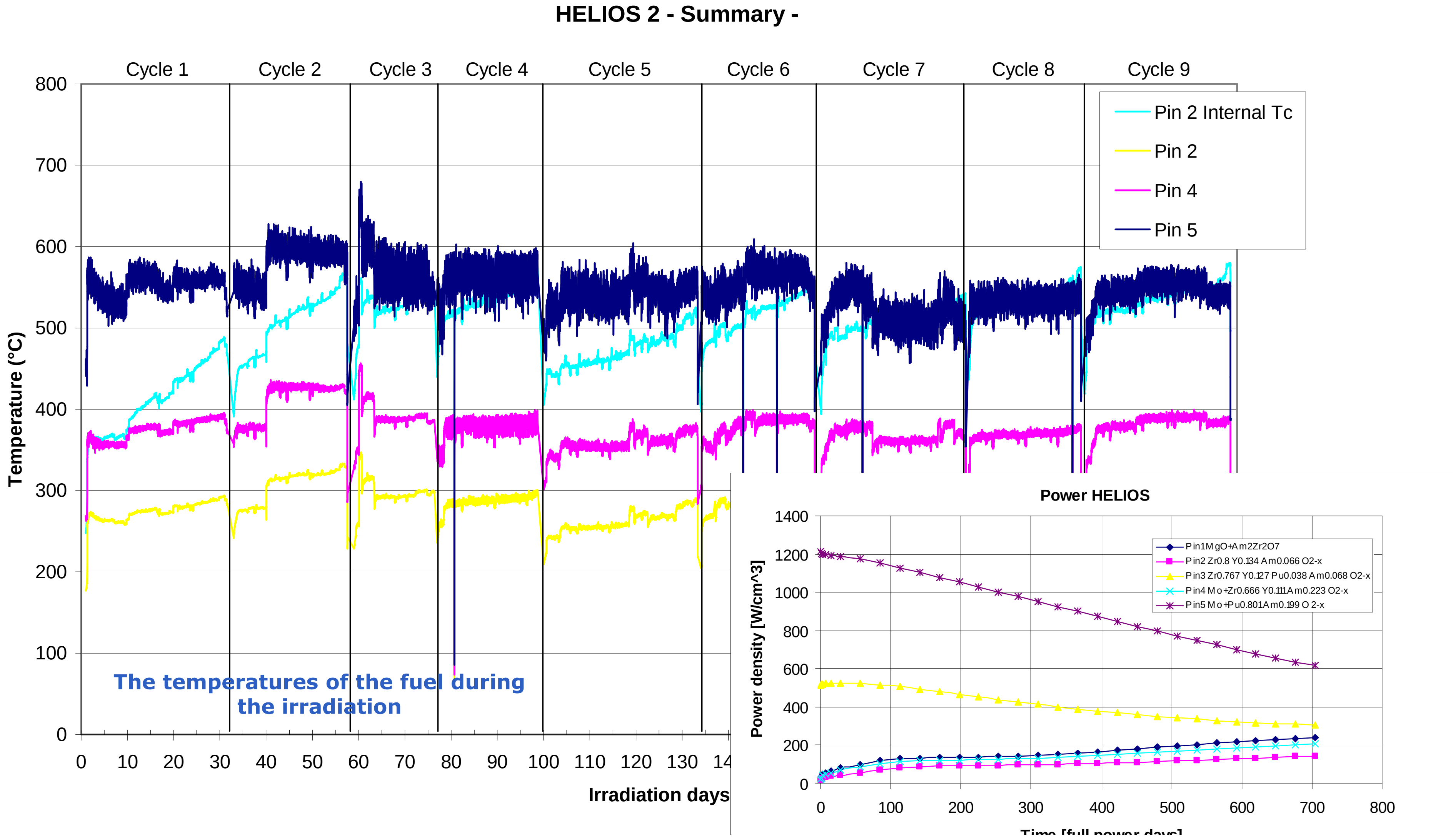
- Pin 1: Am compounds dispersed in a matrix of MgO with tailored open porosity.
- Pin 2 & 3: Am incorporated in a crystal lattice of inert matrix such a zirconia, with and without Pu.
- Pin 4 & 5: Spherical shape particles embedded in an inert matrix (Mo) with and without Pu.



HELIOS irradiation test

- **During Irradiation**

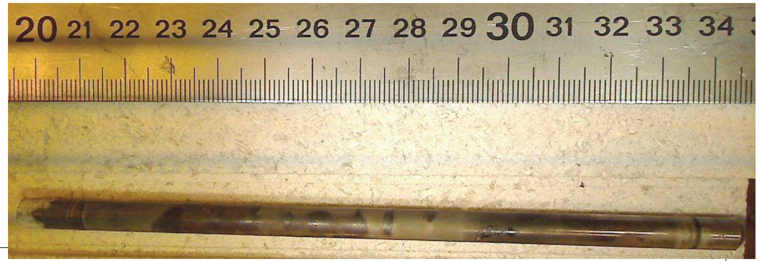


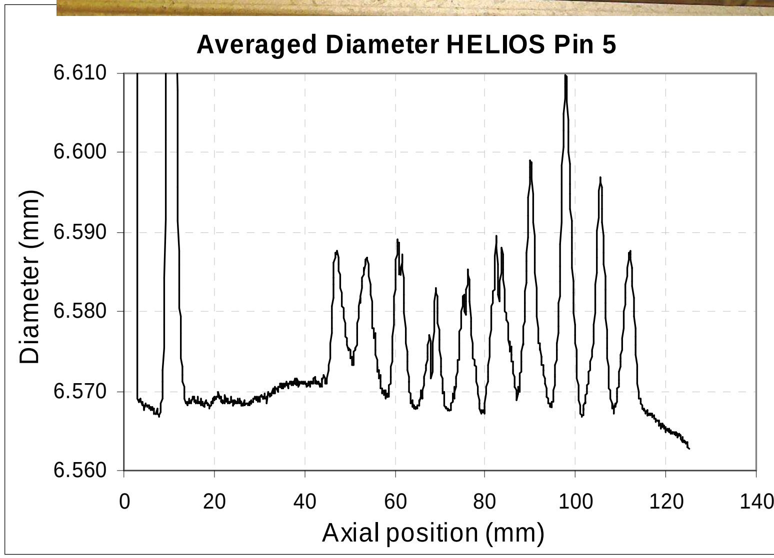


HELIOS irradiation test

- **Post Irradiation Examination (PIE)**

Non Destructive Tests (NDT)





	Before irr.	After irr.	Error margin	(%)
Pin 1	61.92	62.6	± 0.25	**1.1**
Pin 2	61.24	60.1	± 0.25	**-1.9**
Pin 3	62.74	60.8	± 0.25	**-3.1**
Pin 4	59.29	59.3	± 0.25	**0.0**
Pin 5	62.55	65.8	± 0.25	**5.2**

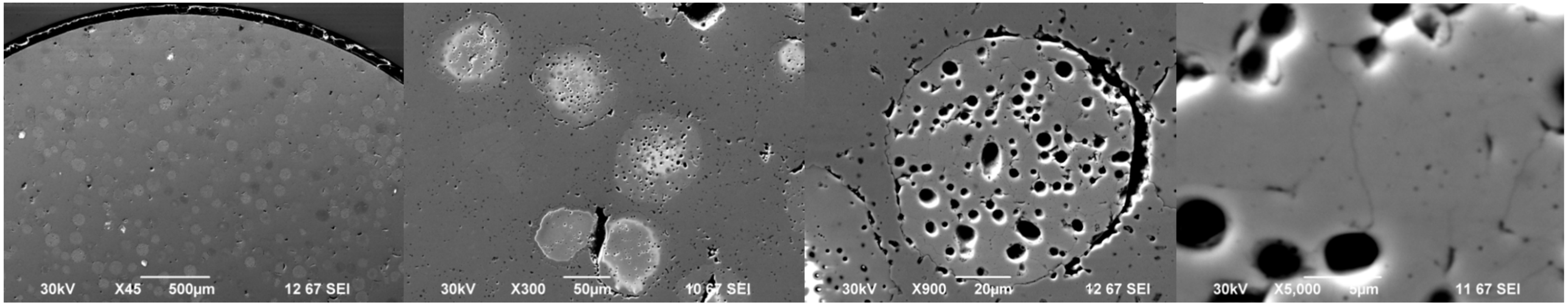
- ✓ Both YSZ pins show slight compaction, possibly due to high porosity;
- ✓ Molybdenum shows large temperature influence.

parameter	units	**caps. #1**	**caps. #2**	**caps. #3**	**caps. #4**	**caps. #5**
experiment		HELIOS I	HELIOS II	HELIOS I	HELIOS II	HELIOS II
material		$(Am_2Zr_2O_7)$+ MgO (porous)	(ZrYAm)O2	(ZrYPuAm)O2	(ZrYAm)O2 +Mo	(PuAm)O2 +Mo
temperature	C	700	500	1150	500	1100
He fiss. released fraction	%	17,24%	2,70%	35.7%	2.5%	28,66%
Kr released fraction	%	9-23%	0,5-1,7%	21.1%	25%	89.2%
Xe released fraction	%	16-28%	0.26-0.45%	27,50%	18%	79.6%

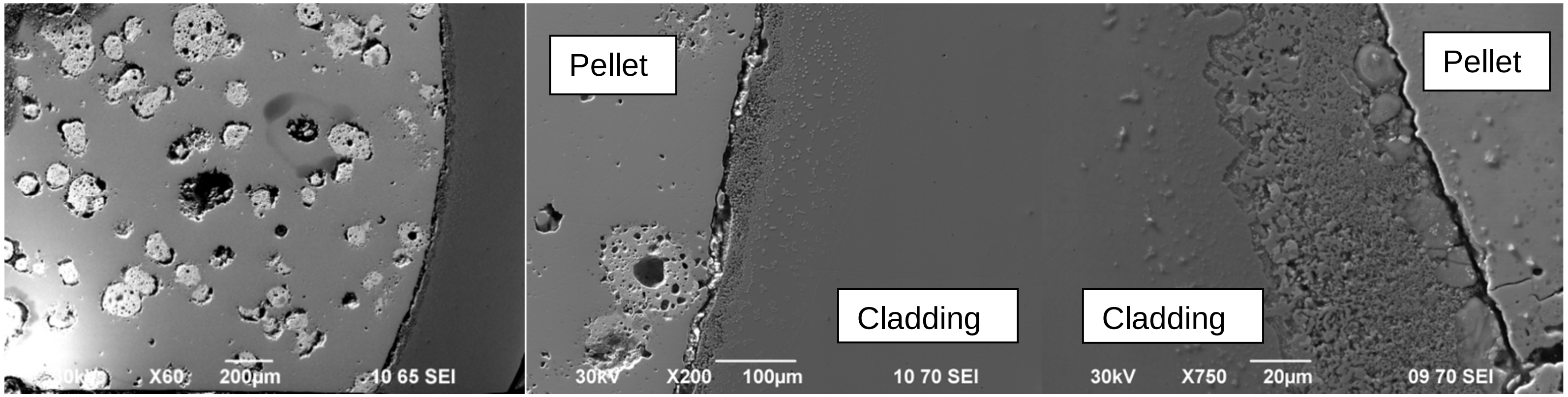
HELIOS irradiation test

- **Post Irradiation Examination (PIE)**
- **Destructive Tests**

PIN 4



PIN 5



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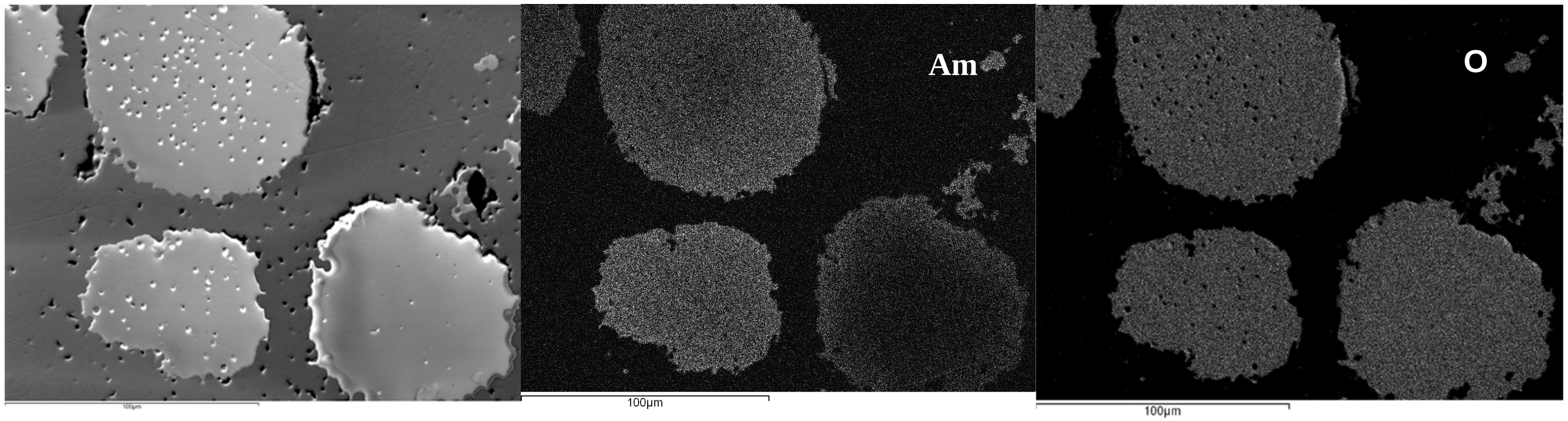
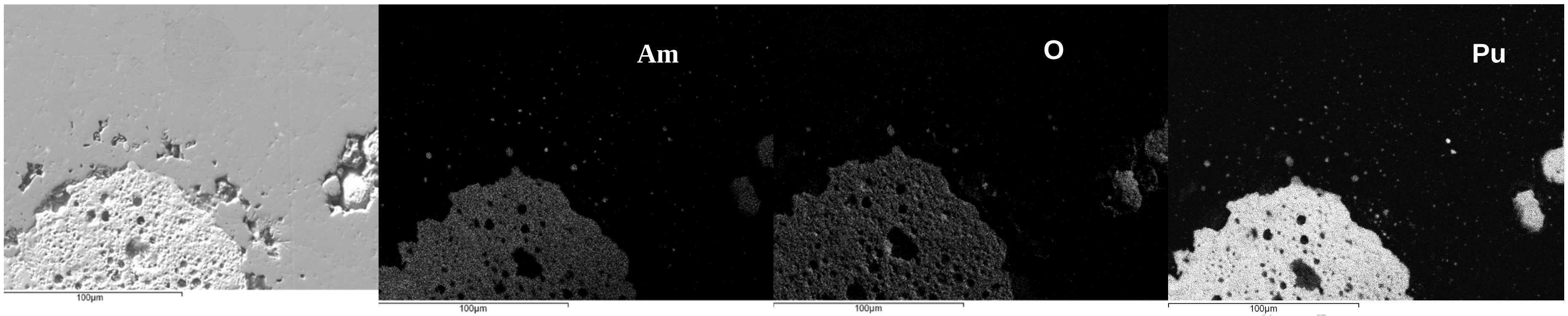
HELIOS irradiation test

- **Post Irradiation Examination (PIE)**
- **Destructive Tests (WDS mapping)**

PIN 4



PIN 5



E. D'Agata et all, Journal of Nuclear Materials 465 (2015) 820-834

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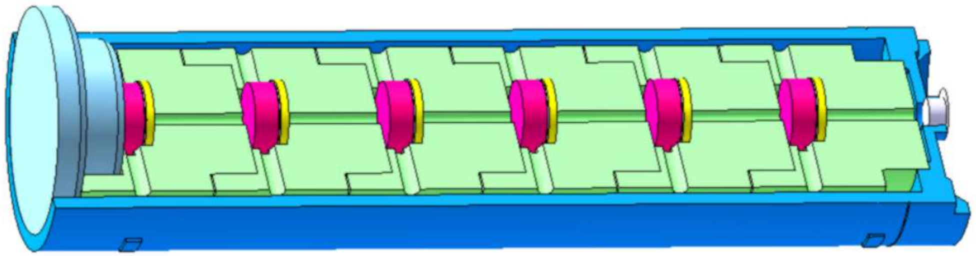
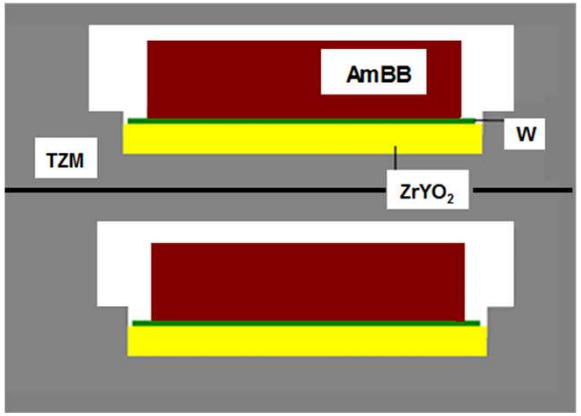
MARIOS irradiation test

- ❑ Purpose of the MARIOS (first) separate-effect irradiation of AmBB
  - ➢ AmBB in SFR: significant He production (Am transmutation) associated with moderate temperatures (500-1500°C)
  - ➢ **The combination (He production / low temperatures) may induce significant fuel swelling**

  → Study of gas (He) release and swelling as a function of temperature and microstructure (level of opened porosity)

- ❑ Small pin innovative design:





Each pin includes 6 small AmBB discs ($Am_{0.15}U_{0.85}O_{1.94}$) and is devoted to one given experimental configuration.

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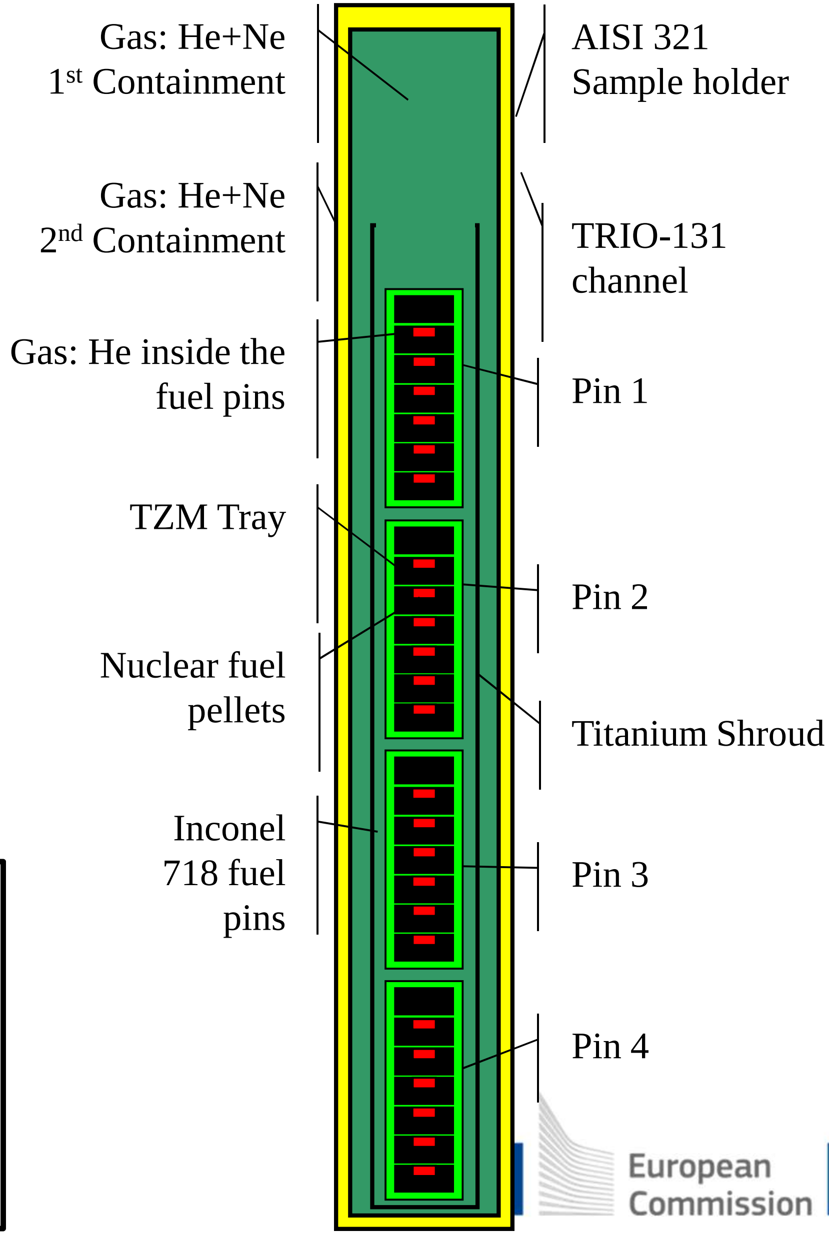
MARIOS irradiation test

- The small pins are set within a Ti shroud containing instrumentation (TCs and FD), the whole being incorporated into a sample holder.
- The sample holder is set itself into a channel of a standard TRIO 131 rig
- Temperature regulation is insured by:
  - gas gaps with adjustable He/Ne composition.
  - a possible axial displacement of the sample holder to optimise the neutron flux profile.



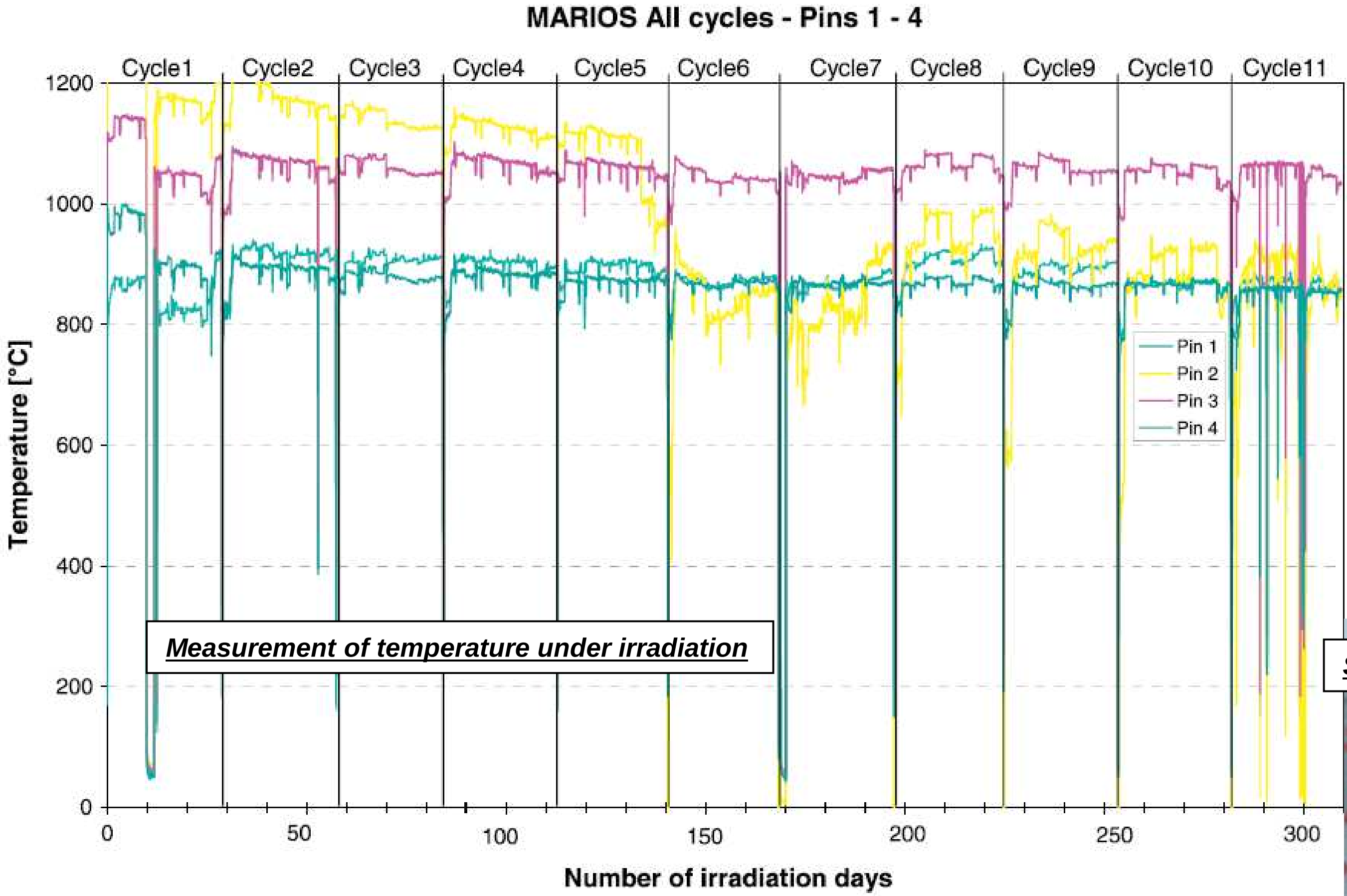
- **304 EFPD irradiation in the HFR**
- **Irradiation conditions of pin n° 3 *(most irradiated)***
  - **→ Transmutation rate: ≈ 55 at%**
  - **→ Fission rate: ≈ 1.5 at%**
  - **→ He production: ≈ 4.7 mg/cm^3**

MARIOS irradiation test

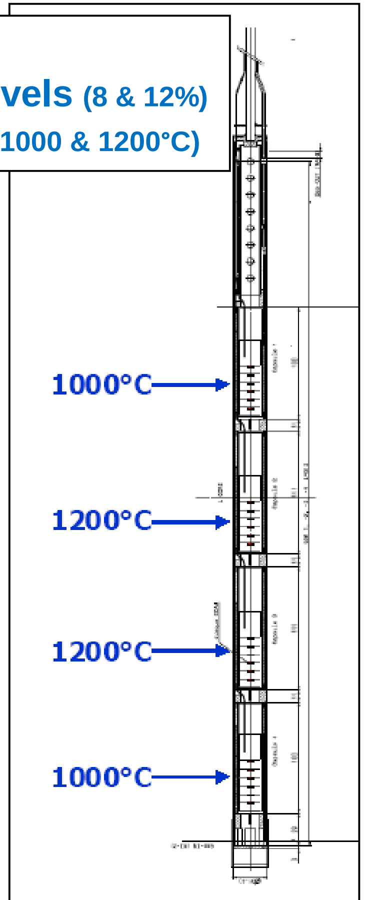
MARIOS separate-effect irradiation in the HFR

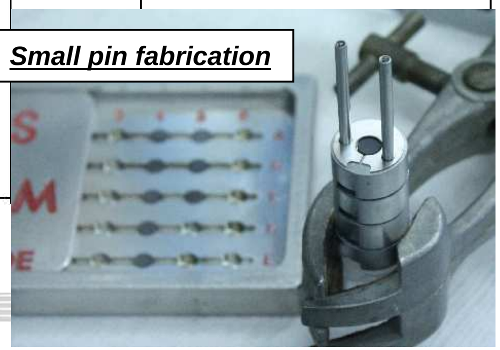
– Fabrication (CEA/NRG) : 2009-2010
– Irradiation (NRG/JRC) : 2011-2012 } Fairfuels
– PIE (NRG/CEA) : 2012-2015→2017 } Pelgrimm

4 pins
2 opened-porosity levels (8 & 12%)
and 2 temperatures (1000 & 1200°C)



Measurement of temperature under irradiation

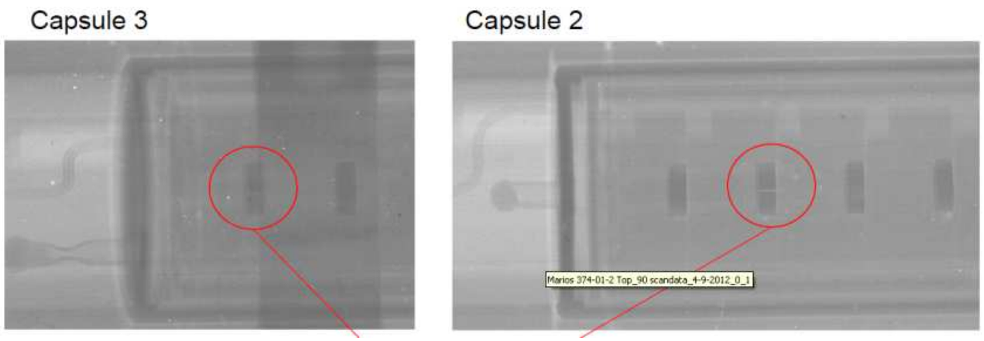




Small pin fabrication

MARIOS irradiation test

- Results of Non-Destructive-Examinations (NDE) performed at HFR by NRG within PELGRIMM



→ Visual inspection and neutron radiography: good behaviour of the pins and the experiment and fragmentation of some discs into 2 pieces (due to thermal gradient).

→ **New and important results obtained through the puncturing of the pins and their gas analysis**

- 100% of the He produced under irradiation is released in the 4 small pins at 1000, 1200 and 1300°C

  He dissolved in the matrix with high diffusion coefficients

- 10, 50 and 90% of the fission gas were respectively released in the small pins at 1000, 1200 and 1300°C

  FG mobility is lower than for He and depends on the temperature

- Destructive Examinations (DE) still to be performed at CEA

- DIAMINO in OSIRIS will give information at 600 & 800°C for both dense and opened-porosity microstructures (Irradiation performed, PIE ongoing).

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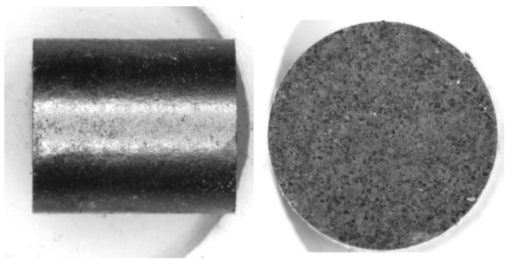
MARINE irradiation test

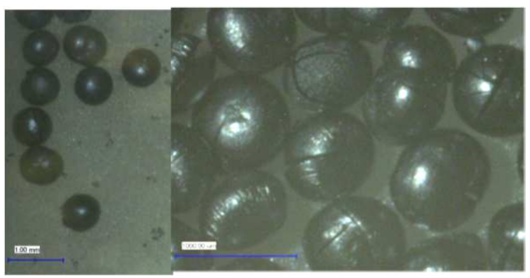
MARINE semi-integral irradiation of AmBB in the HFR

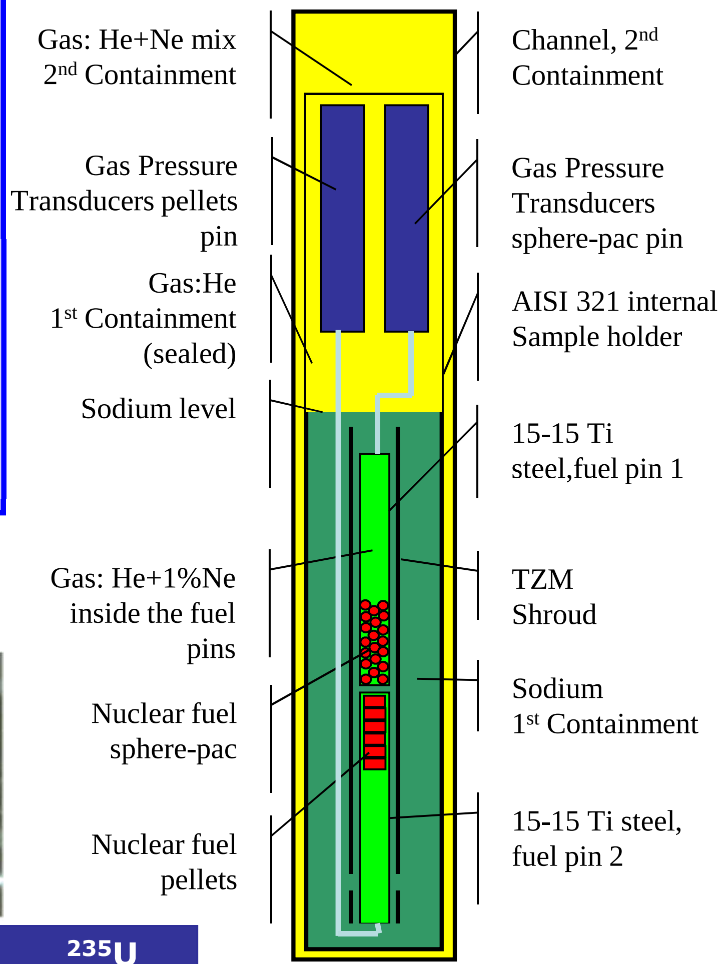
'MA heterogeneous Recycle semi-Integral Experiment'

- Irradiation specifications (CEA) : 2012
- Irradiation design (JRC-NRG) : 2013-2015
- Fuel fabrication by means of Sol/Gel & spherule metallurgy (JRC) : 2013-2014
- Irradiation (JRC-NRG) : 12/2015-5/2017
- PIE: To be defined

(PELGRIMM project)







Pin Nr.	Composition	Isotopic composition	Fuel Density [g cm^{-3}]	^{241}Am contents [g]	^{238}U contents [g]	^{235}U contents [g]
1 Spheres	$U_{0.87}Am_{0.13}O_{1.935}$	UO_2 + ^{241}Am	6.13	1.08	7.152	0.051
2 Pellets	$U_{0.86}Am_{0.14}O_{1.93}$	UO_2 + ^{241}Am	9.45	1.58	10.017	0.072

MARINE irradiation test

- Issue of irradiating AmBB MTR / SFR : High absorption cross-section of Am in thermal neutron flux
  - → Speed-up of transmutation phenomena in MTR - higher volume power and He production rate
  - - higher 'transmutation/fission' ratio

→ Lowering of the irradiation duration (2000 EFPD for reference AmBB ≈ 6~7 years in SFR!)

→ Lowering of pellet diameter to reach LP (beneficial regarding Am needed for the experiment)

→ Speed-up of He production rate & Higher transmutation/fission ratio

→ Radial flux and LP depression in the pellet

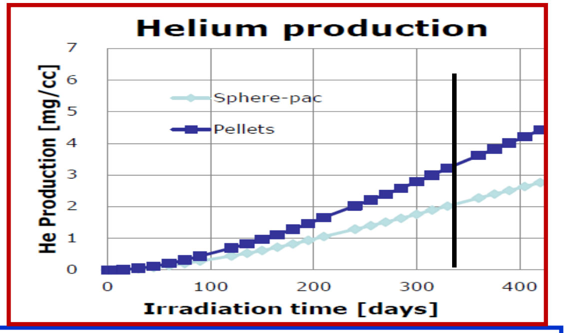
- <u>MARINE irradiation conditions in the HFR</u>

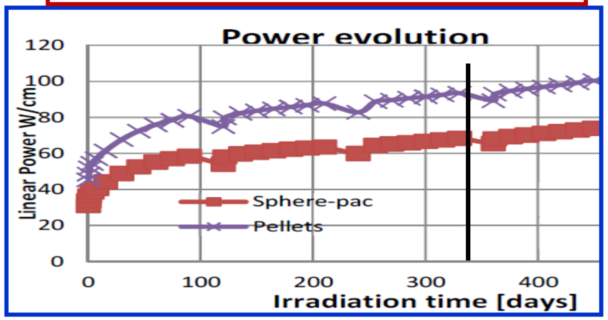
→ 336 EFPD in HFR (vs 2000 EFPD in SFR)

→ Phenix cladding & $Ø_{pellet}$: 5.35 mm (vs 7.34 mm in SFR V2b core)

→ Pellet LP and temperature in acceptable accordance with targeted values

→ Acceleration factor for He production: ≈4.0

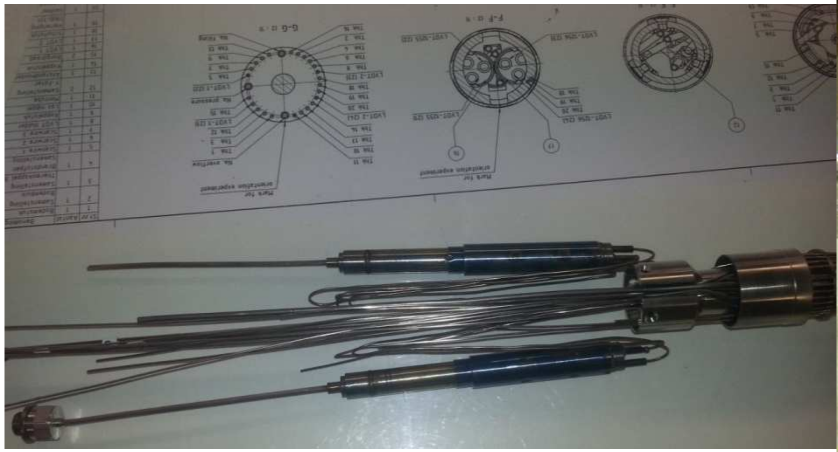


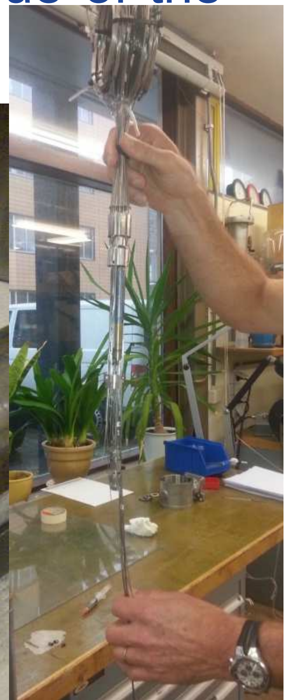


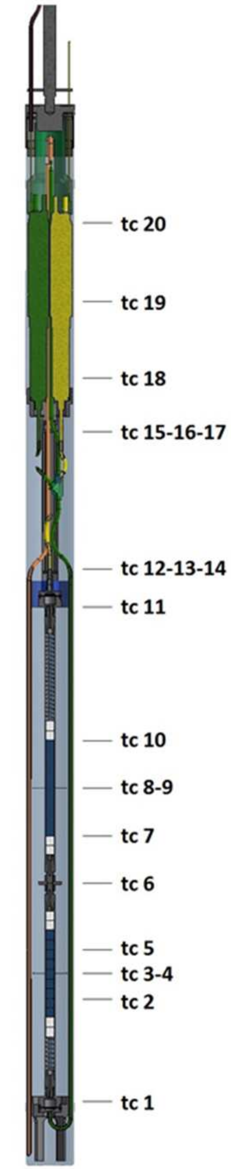
MARINE irradiation test

Instrumentation:

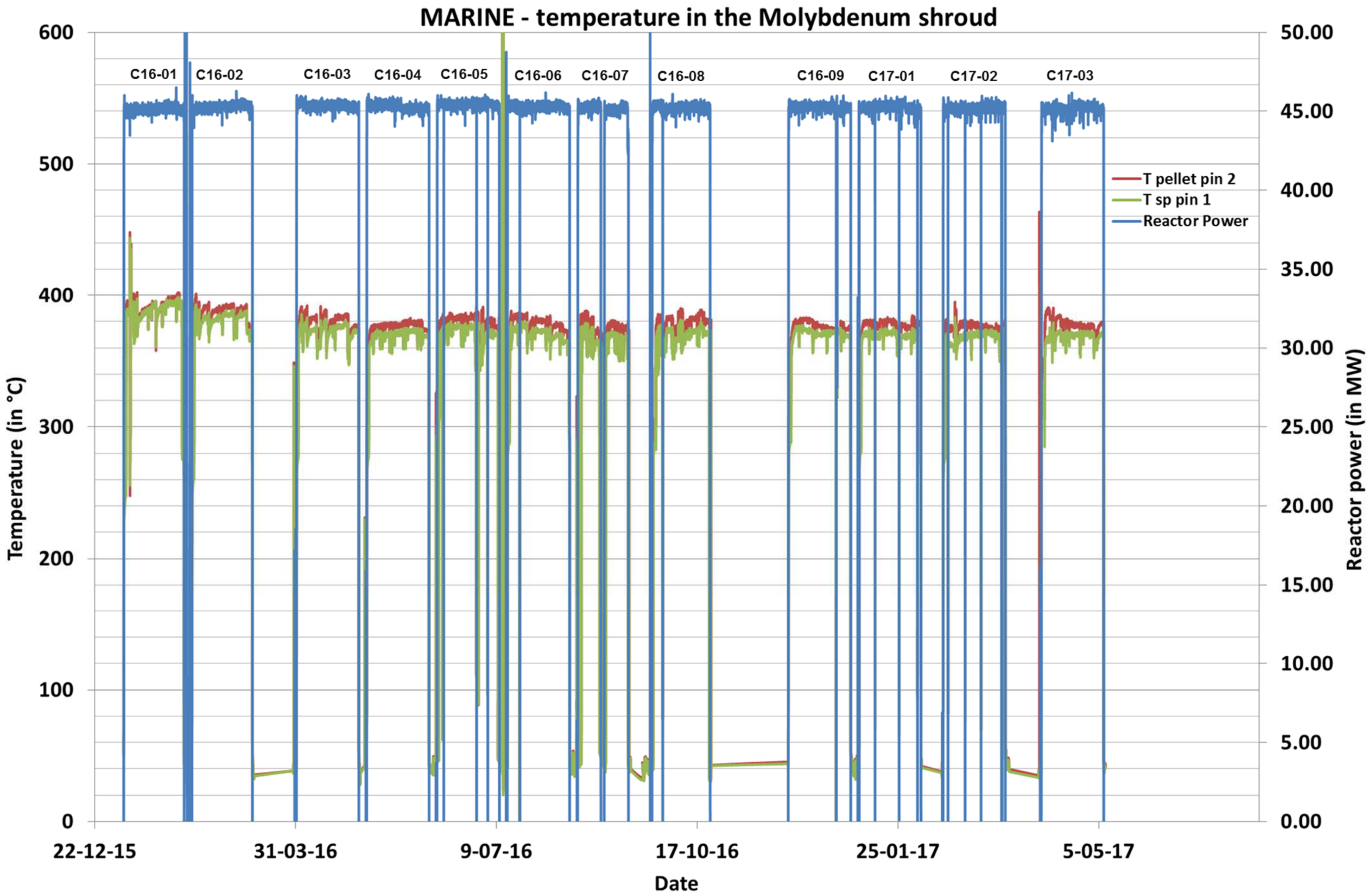
- **20 Thermocouples**: To measure the temperature in the experiment.
- **6 Dosimeter:** To measure the integrated fluence/spectrum.
- **2 Pressure transducers:** To monitor the pressure buildup inside the pins.
- **4 Gamma scan wires**: To get an axial integrated value of the neutron flux





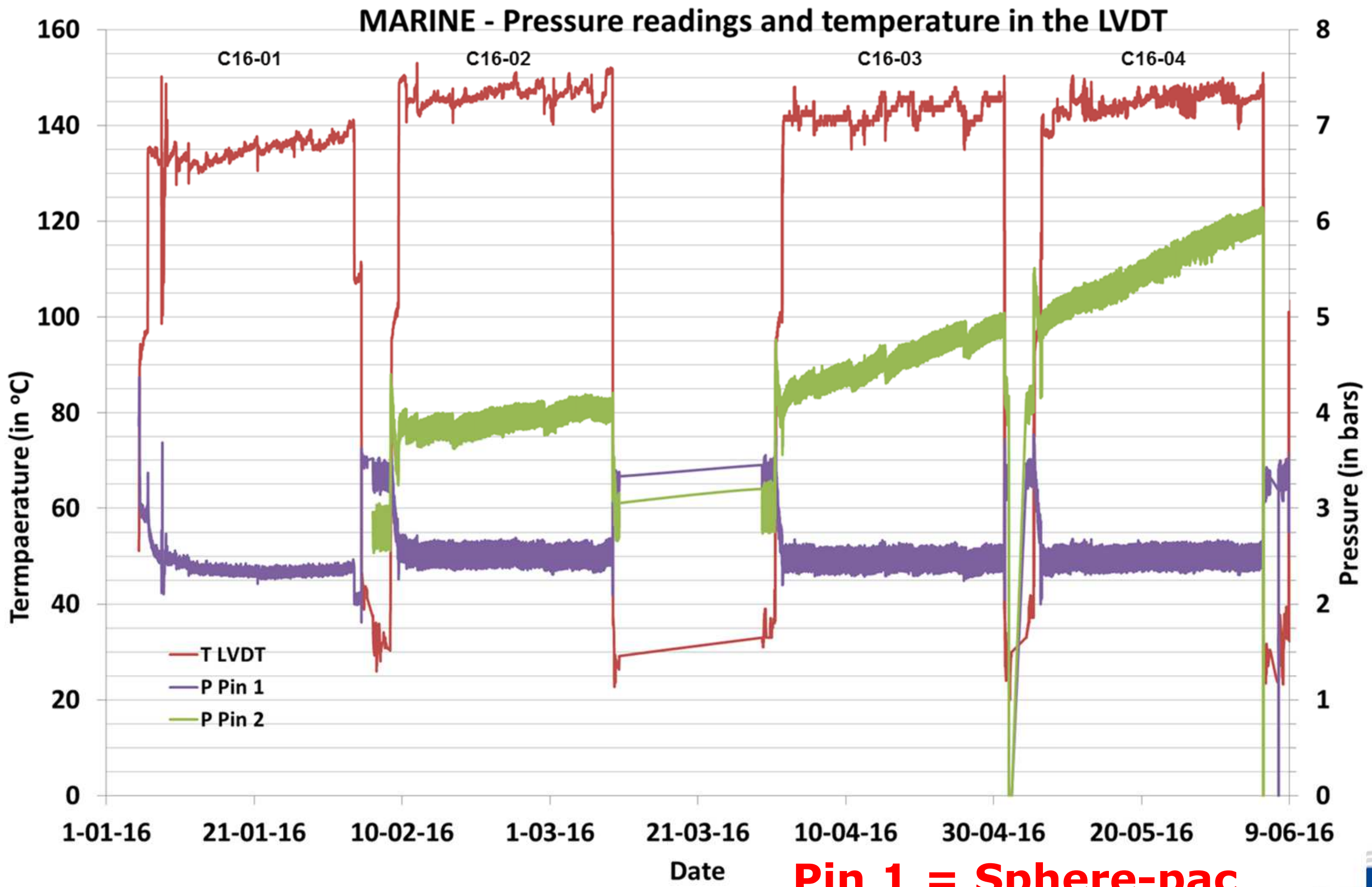


MARINE irradiation test



MARINE irradiation test

Measured Items



Pin 1 = Sphere-pac
Pin 2 = Pellet

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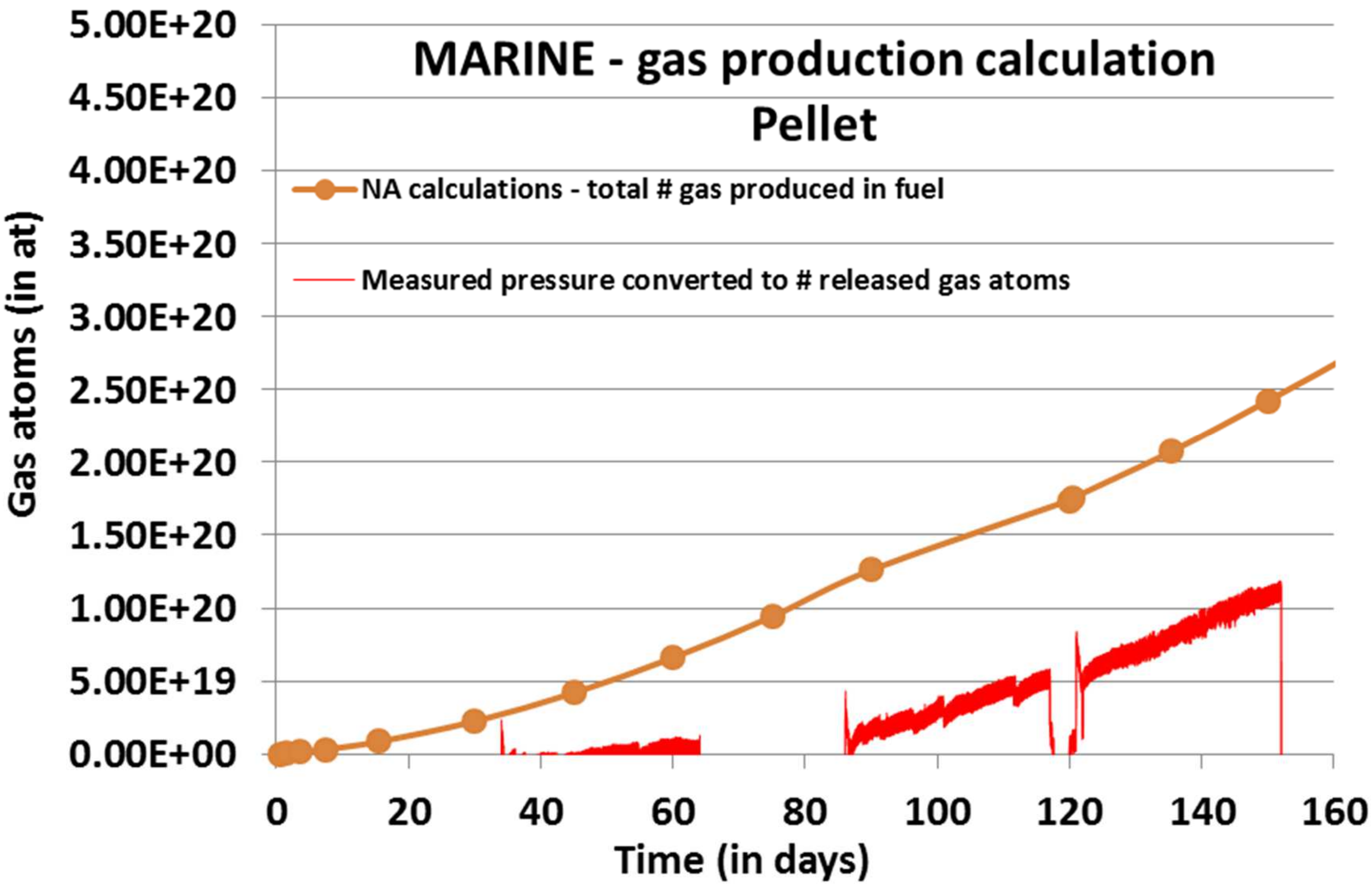
MARINE irradiation test

Measured Items vs Calculated Items



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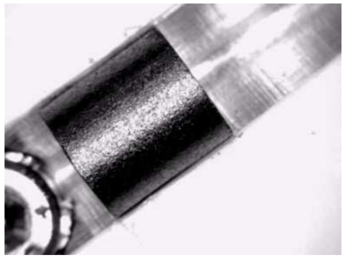
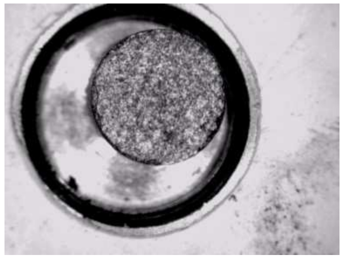
SPHERE irradiation test

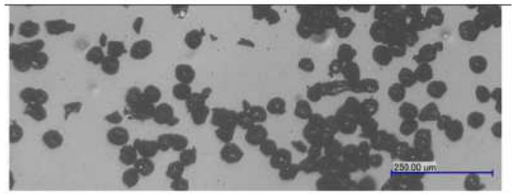
SPHERE semi-integral irradiation of MADF in the HFR (FAIRFUELS & PELGRIMM European projects)

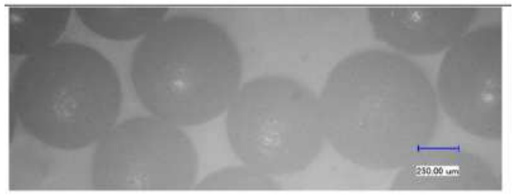
– Irradiation design (JRC-NRG): 2011-2013
– Fuel fabrication (JRC): 2011-2012
– Irradiation (NRG): 2013-2015
– PIE (JRC-NRG): 2015-2017

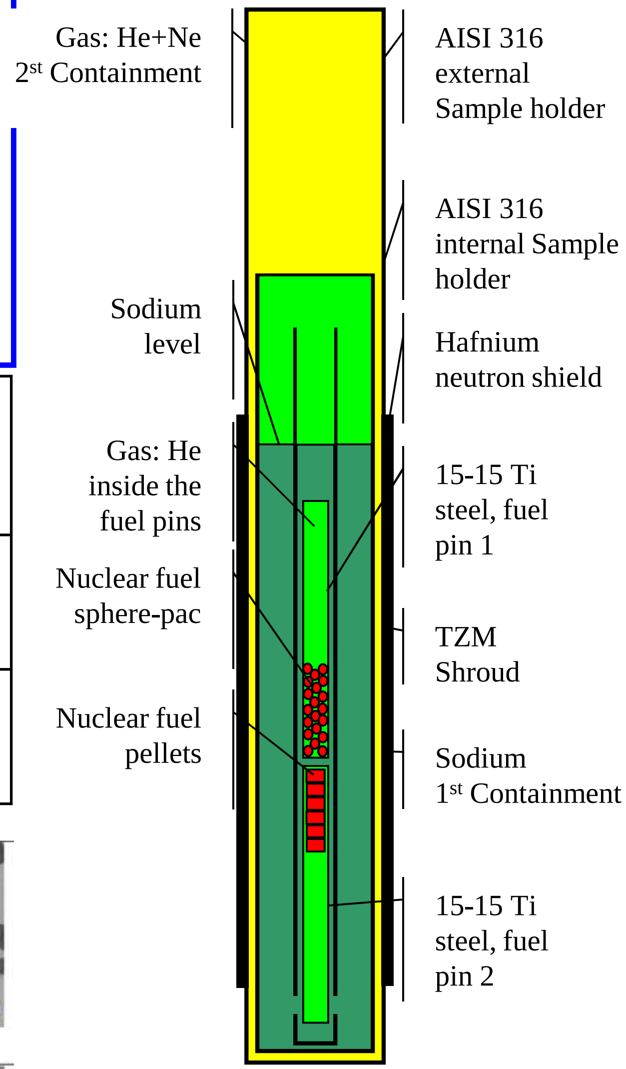
Pin Nr.	Composition	Isotopic composition	Fuel Density [g cm^{-3}]	^{241}Am contents [g]	^{238}U contents [g]	^{239}Pu contents [g]
1 Spheres	$U_{0.75}Pu_{0.22}Am_{0.034}O_{2-x}$	MOX + ^{241}Am	8.33*	0.320	7.167	1.869
2 Pellets	$U_{0.76}Pu_{0.2}Am_{0.03}O_{2-x}$	MOX + ^{241}Am	10.393 9.423*	0.388	10.192	2.442











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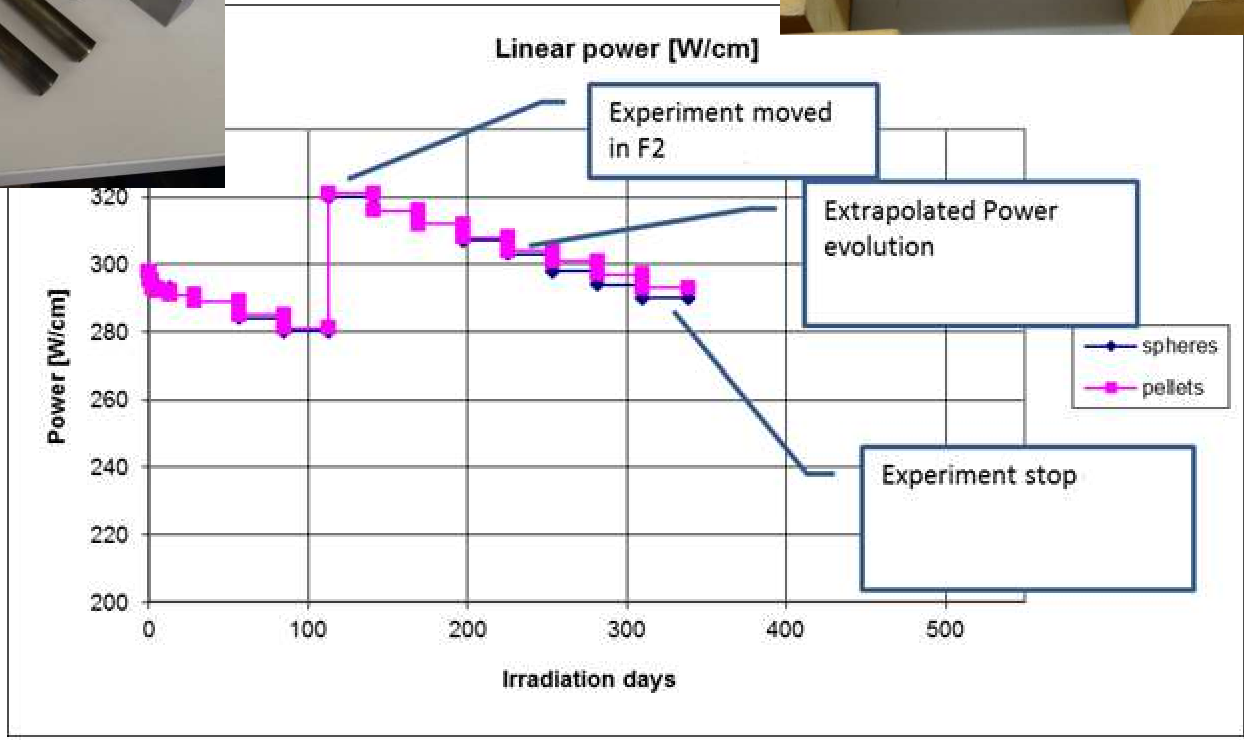
SPHERE irradiation test

- 2 small pins $(U,Pu,Am)O_{2-x}$ with ≈ 20% Pu & 3% Am;
- Comparison of pellet & spherepack fuel behaviour at medium BU;
- Same LP and cladding temperature for the two pins;
- **Fabrication by means of Sol/Gel & spherule metallurgy (JRC).**

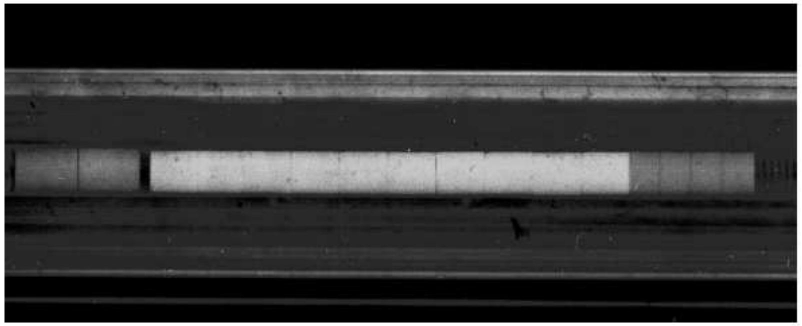


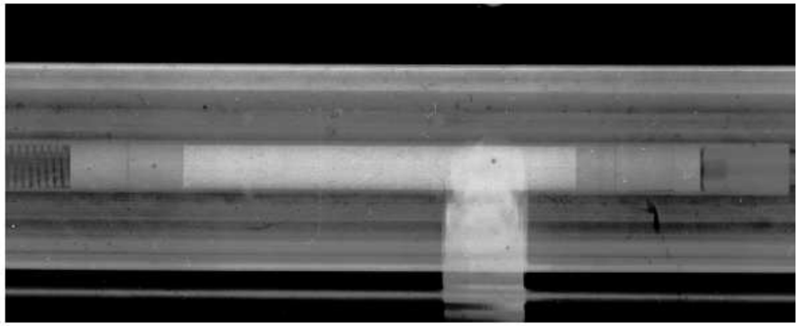
SPHERE irradiation test

❑ SPHERE Irradiation conditions in the HFR

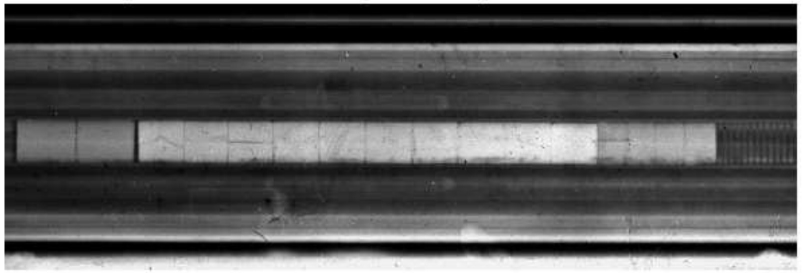
- The first cycle was performed in 03/2013 followed by a neutron radiogram
- Pursuing of the experiment between 02/2014 & 03/2015 → 295 irradiation days
- LP evolution BoL / EoL → 320 / 280 W/cm with position changes at the end of the irradiation
- Fuel max temperature BoL / EoL → 2250 / 2100 °C; BU ~4.5- 5.4 % FIMA

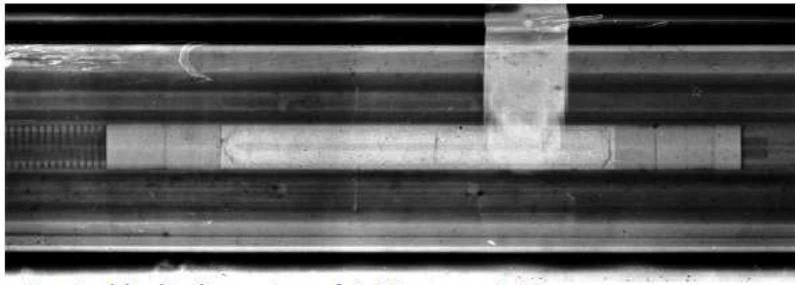
Before irradiation





After 1 cycle of irradiation (~28FPD)

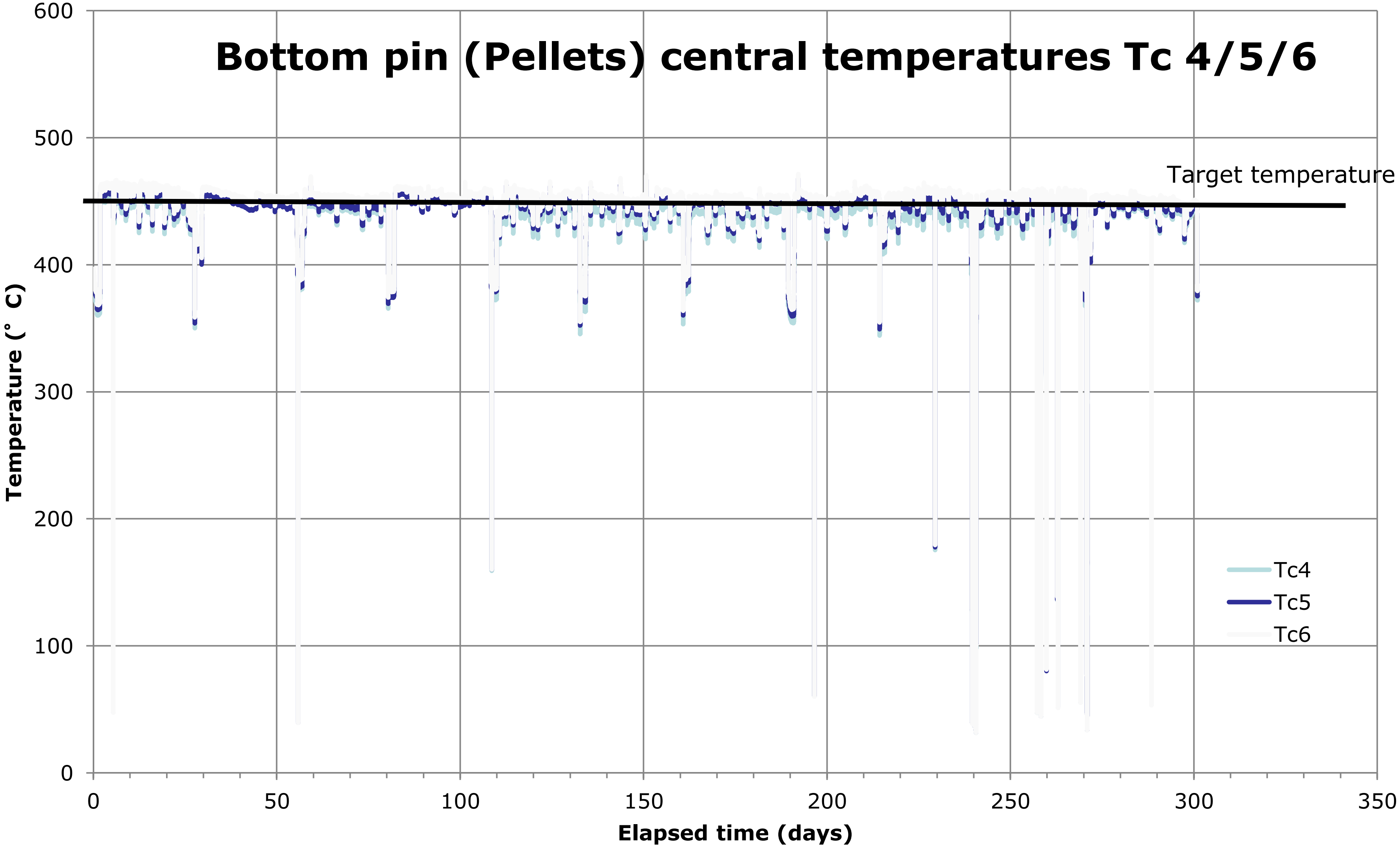




Central hole diameter of 1.05mm ± 0.09mm

Fuel restructuring after 1st cycle

SPHERE irradiation test



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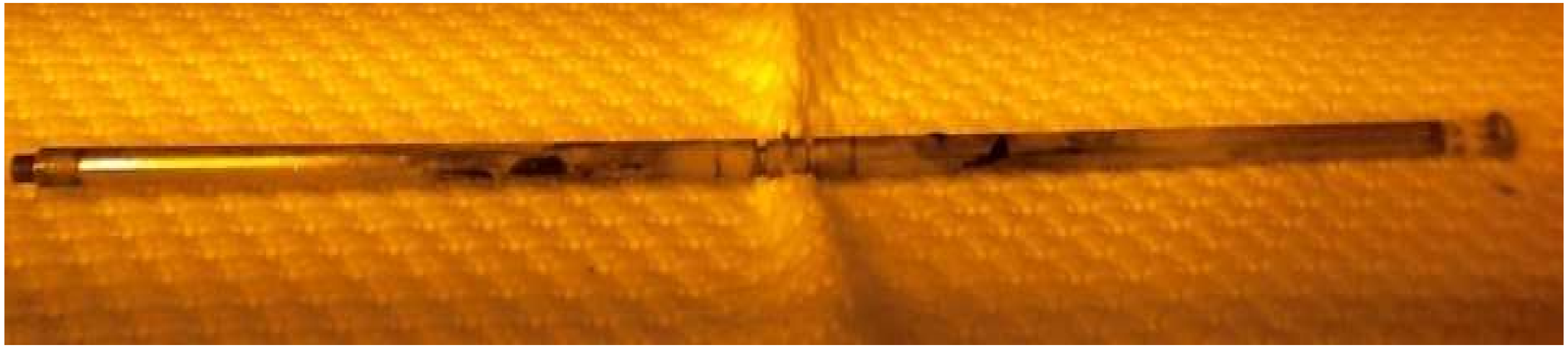
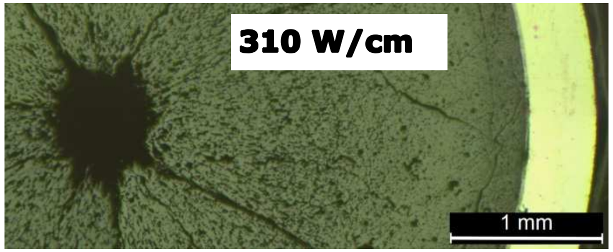
SPHERE irradiation test

- **Post Irradiation Examination:**



- **Dismantling and PIE at NRG**



- **Macroscopic image of restructured pellet (left) and Sphere-Pac (right) fuel sample**

Conclusions 1/2

- **Irradiation experiments are mandatory for existing fuel improvement and new fuel development.**
- **Irradiation experiments are often technically challenging**
  - → **Multi-disciplinary design;**
  - → **Fuel fabrication and transport;**
  - → **Irradiation and instrumentation;**
  - → **PIE with innovative techniques;**
  - → **Working performed in Hotlab facilities;**
- **Irradiation experiments are particularly suitable to collaborative working between different institutes**
  - → **Well-defined objectives;**
  - → **Well-defined tasks that can be easily shared between different contributors;**
  - → **Gain of knowledge for the contributing institutes through information exchanges;**
  - → **Cost-sharing between institutes on relatively expensive experiment projects;**

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Conclusions 2/2

- Presentation of the main features of HELIOS, MARIOS, MARINE and SPHERE experiments.
- **Burn Minor Actinides in Fast Reactor is a viable solution to reduce nuclear waste amount.**
- SPHERE-PAC fuel is promising from fabrication and irradiation point of view.
- The most efficient way (opinion of the presenter) to burn MA in fast reactor is using Heterogeneous Recycle
  - → Higher amount of MA burned (≅ 20%).
  - → In the blanket of FR.
  - → Safety studies didn't have point out problems (opposite to Homogeneous)
- There are still some issues to be tested
  - → Behavior of MA fuel under transient condition
  - → Optimization of irradiation temperature in FR:
    - ❖ High temperature → low swelling → high gas release → need plenum space to accommodate the gas released.
    - ❖ Low temperature → High swelling → the swelling need to be taken into account
  - → Behavior of MA fuel under high Burn-up conditions

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Thank you for your attention...

QUESTIONS?

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