SFR fuels : fundamentals and properties

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ESFR-SMART WORKSHOP,
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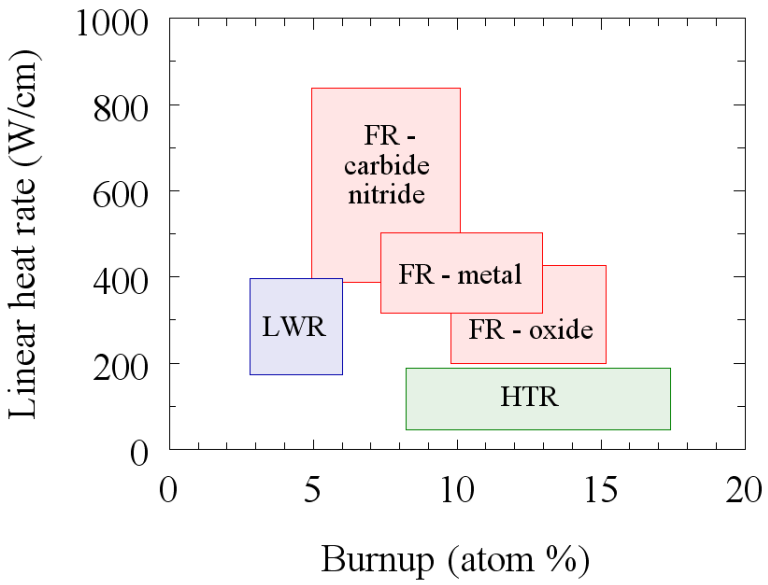
	Gen II/III LWR	*SCWR*	**SFR**	**LFR**	**ADS**	**GFR**	**VHTR**	**MSR**
Fuel +MA	*UO_2, MOX Th-MOX*	*UO_2, MOX Th-MOX*	**$UPuO_2$ UPuZr UPuN UPuC**	**$UPuO_2$ UPuN**	U free fuel, Inert Matrix Fuel	**$UPuO_2$ UPuC**	UO_2,UCO PuO2 $(Zr,Y,Pu)O_2$	$LiF-ThF_4-UF_4$
Cladding	*Zr alloy*	*F/M steel*	**15/15Ticw T91 ODS**	**T91**	T91	**SiC-SiCf**	iPyC/SiC/o PyC	
Liner	-	-	**-**	**-**	-	**W W/Re**	Buf Carbon	Structures
Fuel form	*Pellet*	*Pellet*	**Pellet (Sphere Pac)**	**Pellet (Sphere Pac)**	Pellet (Sphere Pac)	**Plate Pin**	Coated Particle	Fluid
Coolant	*Water*	*Water*	**Na**	**Pb**	Pb or Pb/Bi	**He**	He	$NaF-NaBF_4$

Operating Conditions in Fast Reactors

- High linear heat rate: 400 to 500 W/cm max
- High fuel temperature 600 to 2400°C for $(U,Pu)O_2$

  ~1000°C for (U,Pu)Zr
- High burnup 130 GWd/t or 15 at %
- Residence time >800 days or >130 dpa



GIF (2019), GEN-IV International Forum, « GIF R&D Outlook for Generation IV Nuclear Energy Systems : 2018 Update, » Version 1.1, June 2019.

Criteria for Choice of Fuel Materials

- Material properties
  - High density of fissile atoms
  - High thermal conductivity and high melting point + high thermal stability

    → High margin to melt

    → No phase transition, no dissociation,
  - High mechanical stability
    - Isotropic expansion, radiation resistant
  - Acceptable chemical compatibility with cladding and coolant: no strong reaction

- Performances for evaluation
  - High burn-up and flexibility towards operation conditions
  - Behaviour during transients & accidents
  - Fuel Cycle :
    - Flexibility towards fuel cycle options (Pu and Minor Actinides management)
    - Cost of fabrication and reprocessing

CONTENT

- **Main features of mixed oxide fuel for advanced reactors**
  - Fuel element design
  - Comparison of $(U,Pu)O_2$ properties under irradiation with the others fuels
  - Characteristics of the material
  - Fuel properties

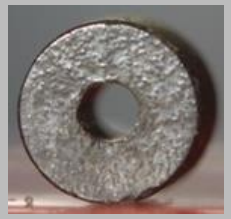
- ***Fuel behaviour under irradiation***
  - *Presented by Dragos Staicu*

- **Fuel element performances, design and qualification**
  - Fuel element performances
  - Improvement in the design and qualification of MOX pins
  - Qualification of fuel performance codes

- **Synthesis & Conclusion**

PART 1 : Main Features of the different fuels for Advanced Reactors

Cadarache facilities:
LEFCA



LECA

Potential fuel designs

e.g. UO_2

Fuel composition
Metal
Oxide
Nitride
Carbide
Fluoride (salt)

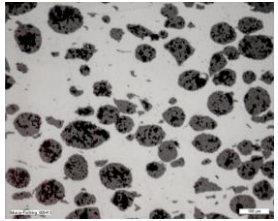
Fuel form
Single phase
Solid solution
Composite

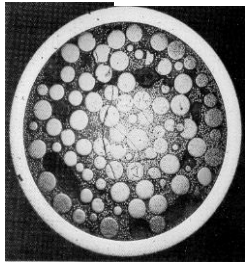
Fuel packing
Pellet
Particle
Liquid

e.g. $(U,Pu)O_2$

e.g. $Mo+UO_2$

Sintered pellets





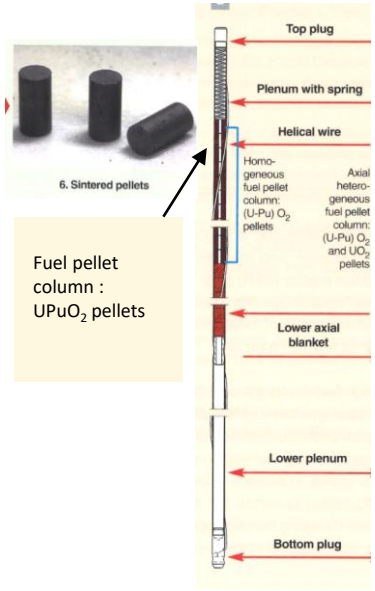
Sphere pack

Y. Guerin International School on Nuclear Engineering , 2009
R. Konings, FJOH 2009
J. Somers, FJOH 2013
T. Wiss, FJOH 2016

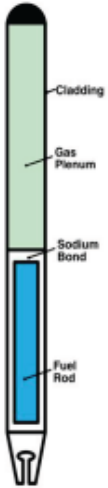
Potential fuel elements (1/2)

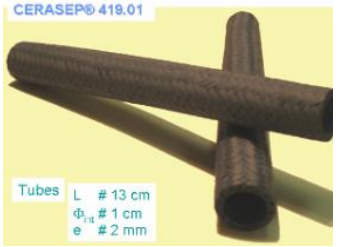
Fuel elements

Standard pin With oxide pellets	Standard pin with metal slug	Pin with innovative clad : composite SiC-SiC$_{fiber}$



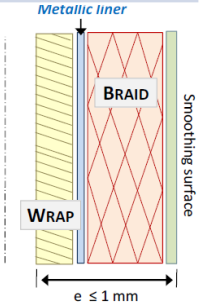
Fuel pellet column : $UPuO_2$ pellets

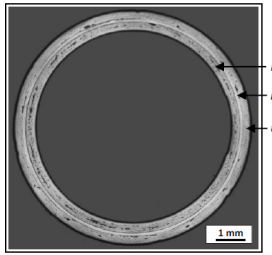




Filament winding
« Inner » SiC/SiC layer

Braiding
« Outer » SiC/SiC layer :
2D ou 3D interlock
One or multilayers
with combiantion of angles
(30, 45, 55 and 70°)

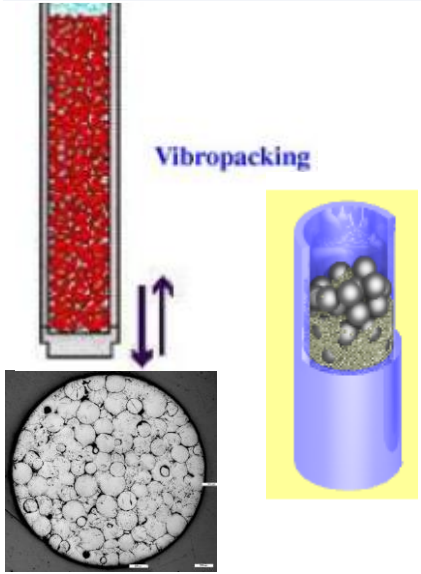


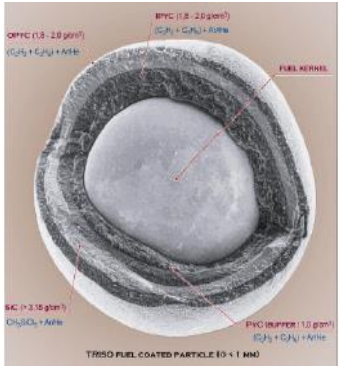


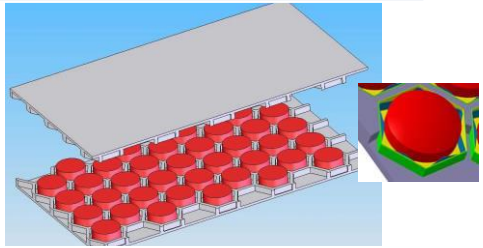
(b)

Potential fuel elements (2/2)

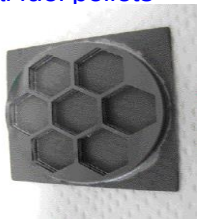
Fuel elements		
Standard pin With vipac fuel or spherepac fuel	Coated particles	Plate fuel







Two ceramic plates close a honeycomb structure containing cylindrical fuel pellets



Comparison of fuel properties during irradiation

Properties	(U0.8Pu0.2)O2	(U0.8 Pu0.2)C	(U0.8Pu0.2)N	U-19Pu-10Zr
Theoretical density, g·cc	11.04	13.58	14.32	15.73
Melting point, K	3083	2750	3070	1400
Thermal conductivity, ($W \cdot m^{-1} \cdot K^{-1}$) at 1000–2000 K	2.6–2.4	18.8–21.2	15.8–20.1	40–40
Crystal structure	Fluoride	Nacl	Nacl	Alfa
Breeding ratio	1.1–1.15	1.2–1.25	1.2–1.25	1.35–1.4
Swelling	Moderate	High	Moderate	High
Handling	Easy	Pyrophoric	Inert	Inert
Compatibility: clad	Average	Carburisation	Good	Eutectics
Compatibility: coolant	Average	Good	Good	Good
Dissolution and reprocessing	Good	Demonstrated	Risk of C14	Amenable for pyro reprocessing
Fabrication/irradiation experience	Large and good	Limited	Very little	Limited

GIF – “Advanced Sodium Fast Reactor (SFR) Fuel Comparison », March 2009.

Metal fuel

Properties	(U0.8Pu0.2)O2	(U0.8 Pu0.2)C	(U0.8Pu0.2)N	U-19Pu-10Zr
Theoretical density, g·cc	11.04	13.58	14.32	15.73
Melting point, K	3083	2750	3070	1400
Thermal conductivity, ($W \cdot m^{-1} \cdot K^{-1}$) at 1000–2000 K	[illegible]	[illegible]	8–20.1	40–40
Crystal structure	[illegible]	[illegible]	Nacl	Alfa
Breeding ratio	[illegible]	[illegible]	2–1.25	1.35–1.4
Swelling	[illegible]	[illegible]	[illegible]oderate	High
Handling	Easy	Pyrophoric	Inert	Inert
Compatibility: clad	Average	Carburisation	Good	Eutectics
Compatibility: coolant	Average	Good	Good	Good
Dissolution and reprocessing	Good	Demonstrated	Risk of C14	Amenable for pyro reprocessing
Fabrication/irradiation experience	Large and good	Limited	Very little	Limited

Low melting temperature
High thermal conductivity
High swelling : large gap + metal bond
Eutectic with clad

Carbide fuel

Properties	(U0.8Pu0.2)O2	(U0.8 Pu0.2)C	(U0.8Pu0.2)N	U-19Pu-10Zr
Theoretical density, g·cc	11.04	13.58		
Melting point, K	3083	2750		
Thermal conductivity, ($W \cdot m^{-1} \cdot K^{-1}$) at 1000–2000 K	2.6–2.4	18.8–21.2		
Crystal structure	Fluoride	Nacl		
Breeding ratio	1.1–1.15	1.2–1.21		
Swelling	Moderate	High		
Handling	Easy	Pyrophoric		
Compatibility: clad	Average	Carburisation	Good	Eutectics
Compatibility: coolant	Average	Good	Good	Good
Dissolution and reprocessing	Good	Demonstrated	Risk of C14	Amenable for pyro reprocessing
Fabrication/irradiation experience	Large and good	Limited	Very little	Limited

High melting temperature + high thermal conductivity:
High margin to melt
Moderate thermal creep
High swelling (to be managed with Na bond or low thermal level or reduced Burn Up)
Fabrication complex, costly

Nitride fuel

Properties	(U0.8Pu0.2)O2	(U0.8 Pu0.2)C	(U0.8Pu0.2)N	U-19Pu-10Zr
Theoretical density, g·cc	11.04	13.58	14.32	15.73
Melting point, K			3070	1400
Thermal conduct ($W \cdot m^{-1} \cdot K^{-1}$) at 1000–2000 K			15.8–20.1	40–40
Crystal structure			Nacl	Alfa
Breeding ratio			1.2–1.25	1.35–1.4
Swelling			Moderate	High
Handling	Easy	Pyrophoric	Inert	Inert
Compatibility: clad	Average	Carburisation	Good	Eutectics
Compatibility: coolant	Average	Good	Good	Good
Dissolution and reprocessing	Good	Demonstrated	Risk of C14	Amenable for pyro reprocessing
Fabrication/irradiation experience	Large and good	Limited	Very little	Limited

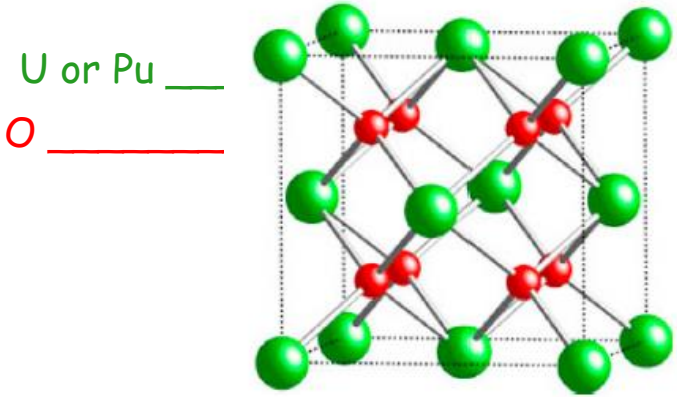
High melting temperature
High margin to melt but possible dissociation at 1800K
High thermal creep (low mechanical interaction with clad)
Moderate swelling

Oxide fuel

Properties	(U0.8Pu0.2)O2	(U0.8 Pu0.2)C	(U0.8Pu0.2)N	U-19Pu-10Zr
Theoretical density, g·cc	11.04	13.58	14.32	15.73
Melting point, K	3083	[illegible]	[illegible]	[illegible]
Thermal conductivity, ($W \cdot m^{-1} \cdot K^{-1}$) at 1000–2000 K	2.6–2.4	18.	[illegible]	[illegible]
Crystal structure	Fluoride	[illegible]	[illegible]	[illegible]
Breeding ratio	1.1–1.15	1.2	[illegible]	[illegible]
Swelling	Moderate	[illegible]	[illegible]	[illegible]
Handling	Easy	Pyrophoric	Inert	Inert
Compatibility: clad	Average	Carburisation	Good	Eutectics
Compatibility: coolant	Average	Good	Good	Good
Dissolution and reprocessing	Good	Demonstrated	Risk of C14	Amenable for pyro reprocessing
Fabrication/irradiation experience	Large and good	Limited	Very little	Limited

High melting temperature
Low thermal conductivity
High margin to melt
High thermal creep (low mechanical interaction with clad)
Low swelling : pin design easier

Structure of mixed oxides $(U_{1-y}Pu_y)O_{2\pm x}$
Face Centred Cubic (fcc) : fluorite type

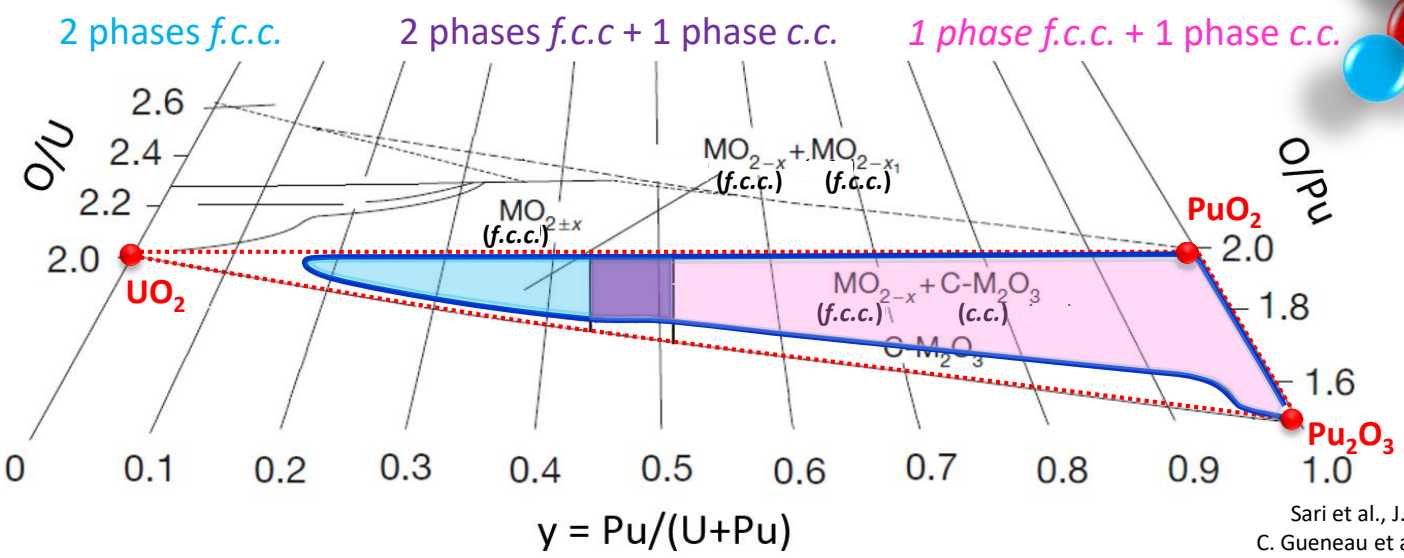


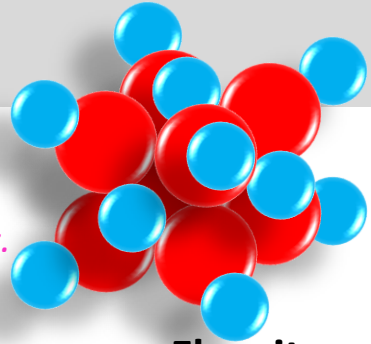
Lattice parameter depends on x and y

- U - Pu substitutions : from 0% to 100% (theoretical)

- Non stoichiometry in actinide oxides
  - $x < 0$: O vacancies or An interstitials (or mixture)
  - $x > 0$: O interstitials or An vacancies (or mixture)

Mixed oxides $(U_{1-y}Pu_y)O_{2\pm x}$





Fluorite (*f.c.c.*)

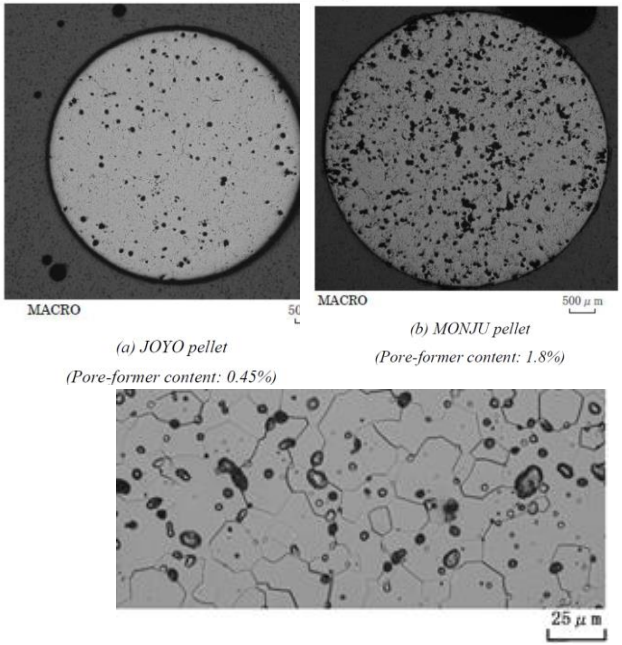
298 K

Sari et al., J. Nucl. Mater.35, 267-277, 1970
C. Gueneau et al. J. Nucl. Mater. 419, 145-167, 2011

- 1,98 < O/M < 2,0 or T>1100K : fcc solid solution
- O/M < 1,98 and T<1100K and Pu>18% with possible phases : $(U,Pu)O_2$, $(U,Pu)O_{2\pm x}$, $(U,Pu)_2O_3$,

MOX fuel : microstructure & fabrication

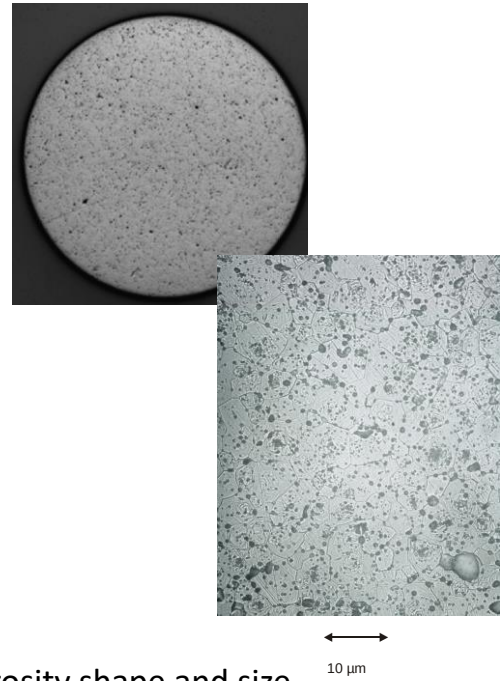
Powder metallurgy
Pore former process
JAEA



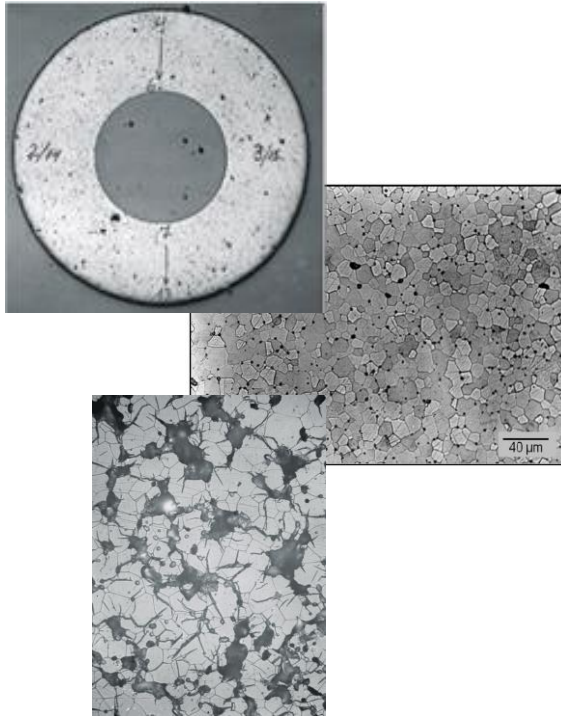
(a) JOYO pellet
(Pore-former content: 0.45%)

(b) MONJU pellet
(Pore-former content: 1.8%)

Powder metallurgy
COCA process
CEA



SOLGEL process
JRC - Karlsruhe



- Microstructure : grain size, density, porosity shape and size
- Microstructure depends on fabrication process

AIEA –TECDOC n°1689 "Design, manufacturing and irradiation behavior of fast reactor fuel".

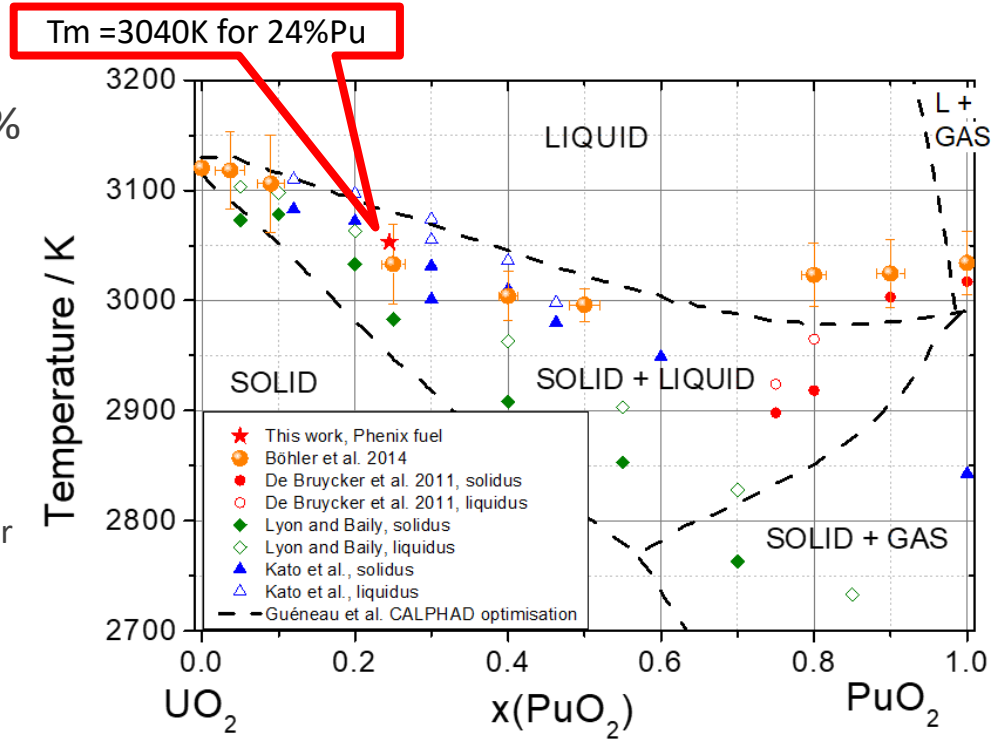
MOX properties :
NEEDS FOR FUEL PERFORMANCE CODES

$(U\text{-}Pu)O_2$ properties / models of interest	Parameters of influence / (Range of interest)						
	Temperature (293 – boiling)	Pu/M ratio (15 – 35%)	O/M ratio (1.94 – 2.00)	Fract. porosity (0 – 40%)	Grain size (4 – 30 µm)	Stress (1 – 100 MPa)	Burn up (0-125 GWd/t)
Lattice parameter	X	X	X				X
Thermal conductivity	X	X	X	X			X
Melting point		X	X				X
Specific heat capacity	X	X	X				X
Enthalpy of fusion		X	X				X
Emissivity	X	X	X				X
Theoretical density	X	X	X				
Thermal expansion	X	X	X				X
Elastic constants	X	X	X	X			
Brittle-to-ductile transition temperature		X	X	X			
Yield stress, ultimate stress	X	X	X	X			
Thermal creep	X	X	X	X	X	X	X
Diffusion / migration of pores, of fission gas, of oxygen, of U, of Pu	X	X	X				
Oxygen potential	X	X	X				X
Grain growth	X			X	X		

New measurements expected

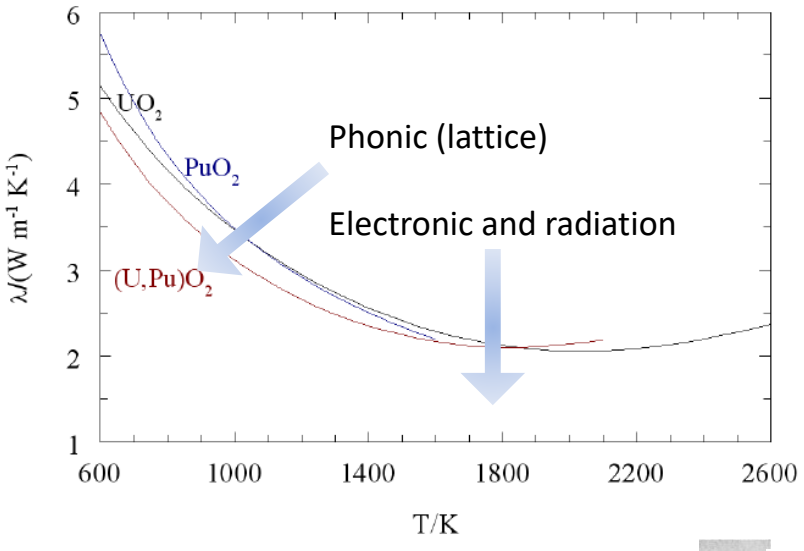
Melting temperature of (U,Pu)O2

- Existence of a minimum around 60-70% Pu content to be confirmed
- Disparity of measurements above 60% of Pu (200 K deviation for PuO_2)
- O / M impact to be evaluated
  - CALPHAD evaluation under estimates solidus.
  - The existing law for melting temperature should be revised following all these and other recent results.
- Needs for additional measurements :
  - → high Pu content
  - → Effect of O/M
  - → Pu% for the lowest $T_{melting}$ (safety analysis)

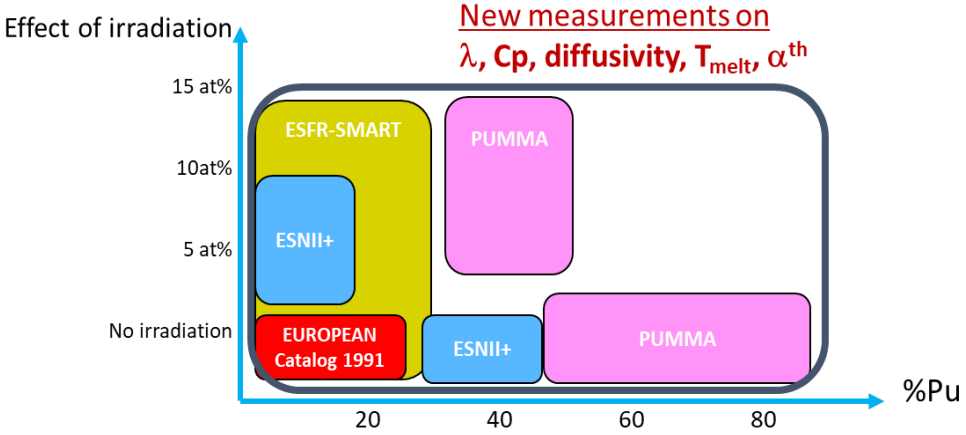


Thermal conductivity of $(U,Pu)O_2$

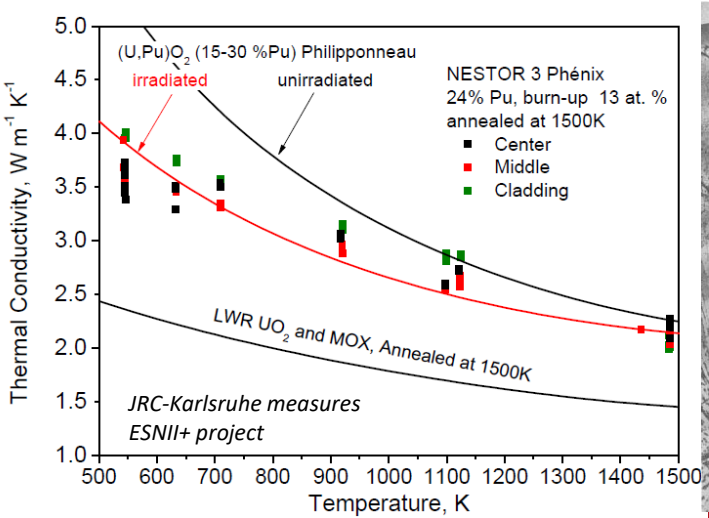
- Strong effect of temperature, O/M, Pu content, density, irradiation :
  - → Discrepancy between the laws of λ
  - → main source of uncertainty on the fuel temperature

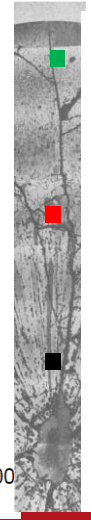


- Intensive European experimental programme:

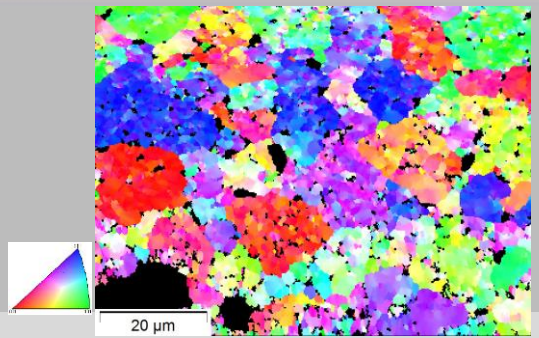


ESNII+ (2013-2017)	ESFR-SMART (2017-2021)	PUMMA (2020-2024)





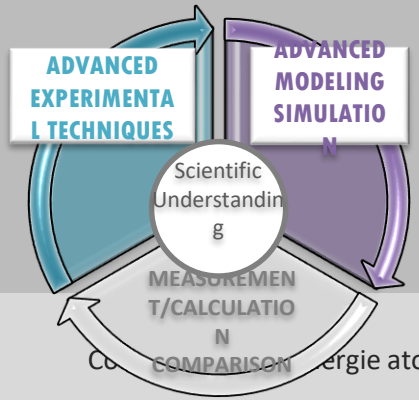
NESTOR fuel irradiated in Phénix (LECA)



PART 2 : Fuel Behaviour Under Irradiation (presented by Dragos STAICU)

SFR MOX driver fuel : main features of the irradiation behaviour

- Microstructure, composition and properties evolution
  - Microstructure evolution early during the irradiation
  - Species transport (actinides and O migration) and oxygen potential evolution
  - Fuel gaseous and solid swelling, coupled with creep
  - Fuel properties evolution depending on : composition, density, microstructure, temperature, burn-up
  - Clad properties : creep, swelling at high burn-up/temperature, embrittlement, loss of mechanical properties under irradiation
- Thermomechanical and thermochemical behaviour
  - Fission gas release : effect of all parameters (T, Burn-up, fuel microstructure,…)
  - Fuel to clad gap closure and Heat transfer in the fuel-clad gap
  - JOG formation and axial transfert
  - Pellet cracking and re-location of the fragments
  - FCMI: threshold of over-power or over-temperature during transients
  - Burn up linked phenomena: FCCI (clad corrosion), JOG composition



PART 3 : Fuel Element Performances, Design and Qualification

Performances of MOX Pins in SFR

- Long experience:
  - Started in BR-5 in 1957 in Russia, Rapsodie in 1967 in France, SEFOR in USA,
  - Then EBR-II and FFTF in the USA, BR-10, BOR-60, BN-350, BN-600 and BN-800 in Russia, the prototype fast reactor (PFR) in the UK, Phenix and Superphenix in France, KNK and SNR-300 in Germany, JOYO and Monju in Japan, FBTR in India and Experimental Fast Reactor (CEFR) in China

Standard MOX fuel		Experimental fuel		
No. of pins irradiated	Burnup reached $MW \cdot d \cdot t^{-1}$	Max. burnup $MW \cdot d \cdot t^{-1}$	Main reactors	Type of fuel pellets
265000	135000	200000	PHÉNIX, PFR, KNK-II	Solid&annular
64000	130000	200000	FFTF	Leading pins
50000	100000	120000	JOYO	Solid
13000	135000	240000	BOR-60	Vibro-pac
1800	100000	-	BN-350	Solid&annular
1500	100000	-	BN-600	Solid&annular

- Performances in recent SFR :

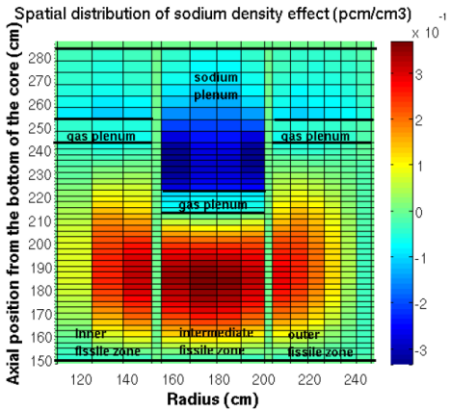
→more than 20 at% (200 GWd.t-1)

→155 dpa

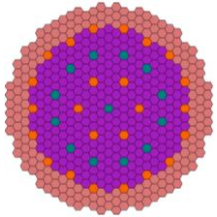
→ 550 W/cm max

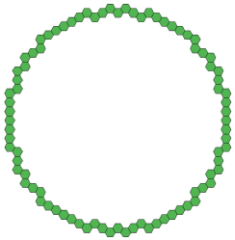
- Fast reactor in operation :
  - BOR60, BN600, BN800 : UOx and MOX, pellet or vi-pack
  - JOYO : MOX, pellet
  - FBTR : carbide and MOX experimental pins and *PFBR : MOX, pellet*
  - CEFR : UOX, pellet

Improvements in the Fuel Element Design



- Improvements on the geometry:
  - Annular pellet (for safety improvement : BOL & transients)
  - large pin diameter
  - Axially heterogeneous pin (safety improvement)
- Large range of composition :
  - Uranium: natural, depleted, reprocessed
  - Plutonium: 15 to 45%, several grades (ex spent LWR-MOX)
  - Minor actinides : transmutation with different ways $(U,Pu,Am)O_{2-x}$, $(U, Am)O_{2-x}$, $(MA,O)_{2-x}$+ inert matrix
- Specifications:
  - Mastered by several processes
  - Adapted to industrial fabrication
  - Responding to safety issues





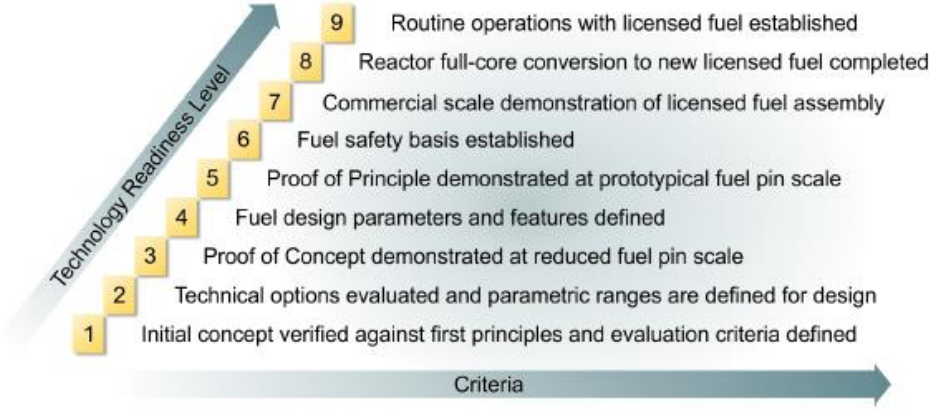
- Objectives :
  - Licensing : authorisation of safety authorities for fuel loading in NPP

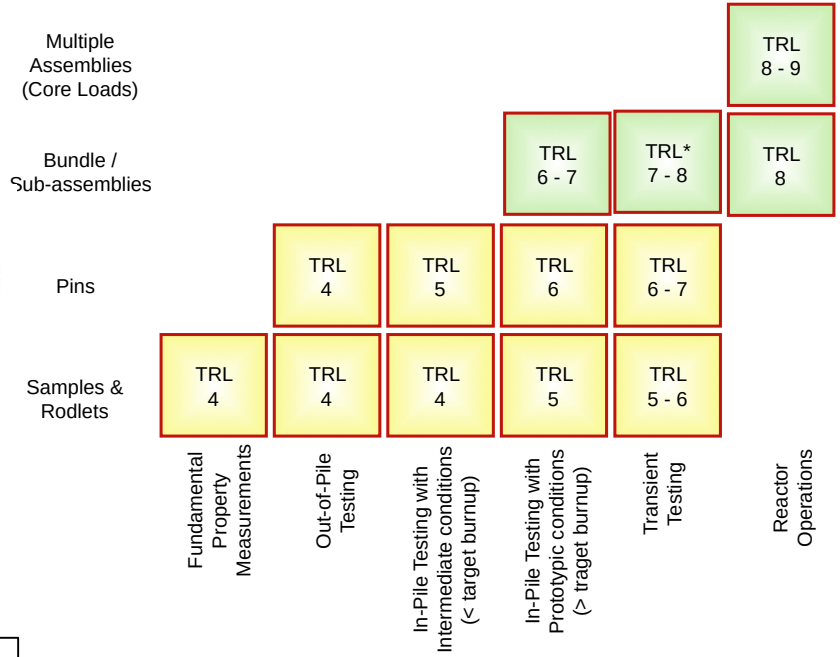
- Requirements:
  - regulatory guidance :
    - higher level safety objectives,
    - qualification of computational tools,
    - uncertainties consistent with Safety Margins

  - regulatory criteria/limits :
    - maintaining cladding integrity, coolable geometry, and limiting radiological consequences
    - fuel failure and degradation mechanisms are identified & controlled

Fuel Element Qualification

- An essential part of fuel qualification is to define a test envelope to cover expected operating, transient, and accident conditions to assess fuel performance and validate fuel performance codes.

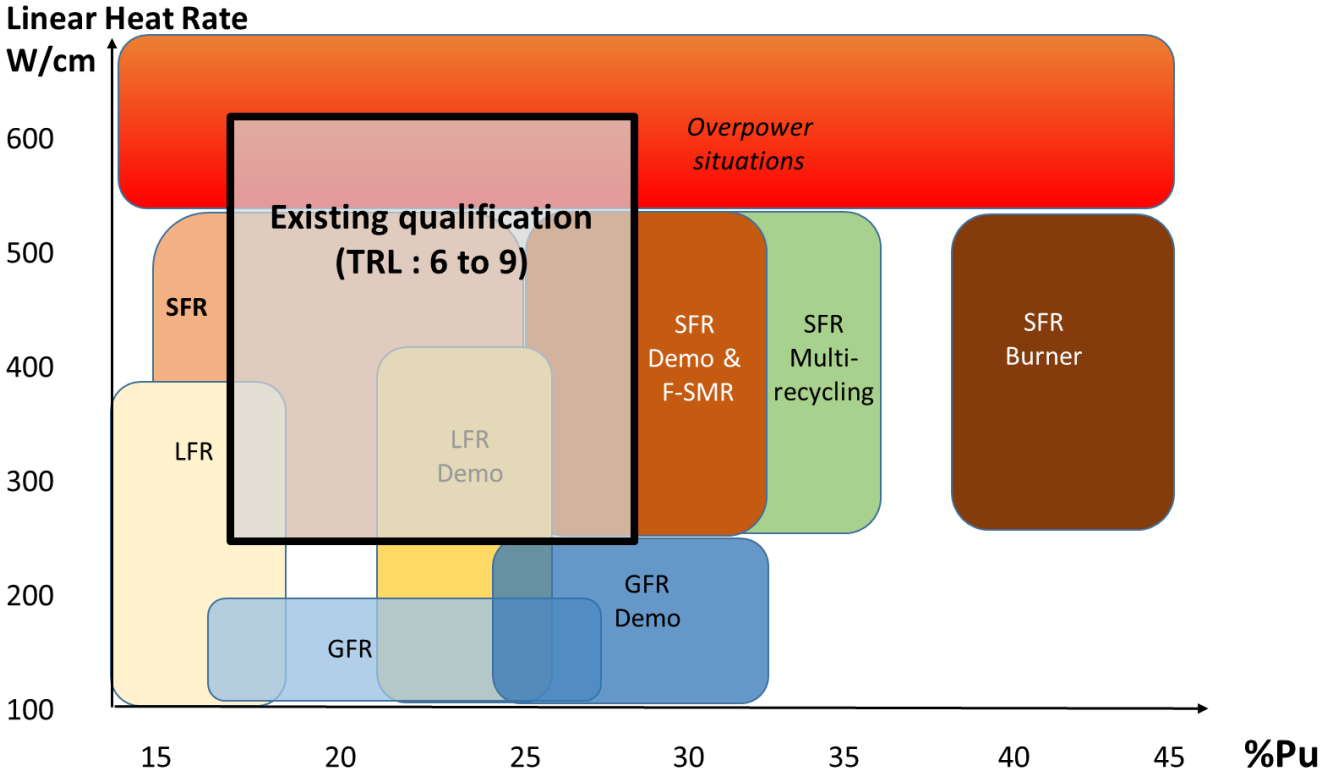




TRL 9 for Phenix type pins (Phenix, SNR300, Joyo) & same geometry with central hole (EBR2, PFR, BN800), SPX type TRL 6-7 for others concepts

D. Crawford et al. , Journal of Nuclear Materials, 371 (2007) 232-242.
OECD – report No. 6895 Dec. 2014. "State-of-the-art Report on Innovative Fuels for Advanced Nuclear Systems".

QUALIFICATION OF MOX FUEL FOR GENIV SYSTEMS



Fuel performance code qualification : ex. platform PLEIADES

with GERMINAL for fast reactor

Material evolution → **Material damage & properties**

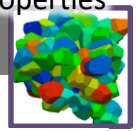
Thermal analysis → **Fuel temperature distribution**

Mechanical calculations → **Mechanical fields**

Physico-chemistry analysis → **Fission gas Location/Migration/ Release**

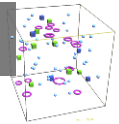
Thermochemistry → **Fission product speciation Thermo-migration Source term**

VER

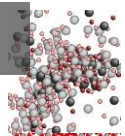


Dislocation Dynamics

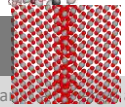
Cluster dynamics

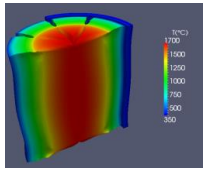


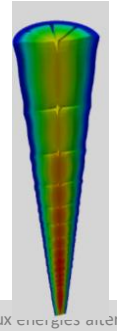
Empirical Potential MD

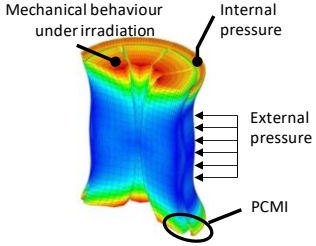


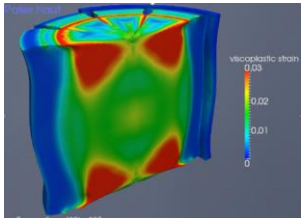
First Principles DFT



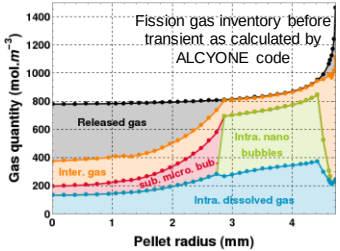


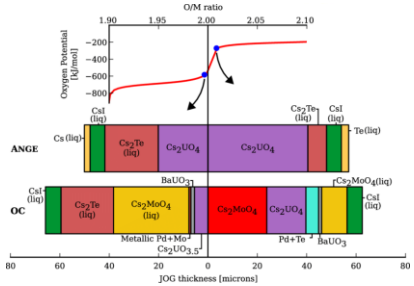






MARGARET/CARACAS

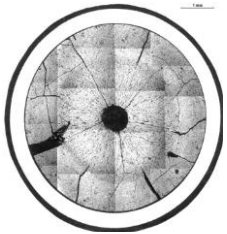




B. Michel et al. chapter 9 in publishing series in Energy, 2021, pages 207-233.
C. Valot, Annual NSC meeting,, 2019. Linking modeling/simulation and experiments for Fuel R&D at CEA
Portelette et al, J. Nuc. Mat., 510, 2018
Bourasseau et al, J. Nuc. Mat., 517, 2019
C. Gueneau, JC. Dumas, Advances in Nuclear Fuel Chemistry,Publishing Series in Energy, 2020, Pages 419-467

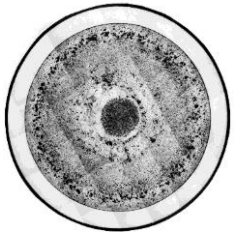
Synthesis & Conclusion

- Oxide fuels
  - Demonstrated a good stability and behaviour under irradiation up to very high burn-ups (20 at.%), limitation is due to clad and wrapper deformation.
  - High creep rate (high temperature) and optimised pin design (smear density) to avoid FCMI
  - Low thermal conductivity compensated by high melting point
  - Compared to metal fuel, lower fuel swelling under irradiation
  - Na reaction to be managed as well as clad corrosion at high burn-up
  - Compatibility with stainless steel cladding
  - Large feed back of safety tests
  - Fuel Performance Codes : numerous and qualified on a set of reliable exp. tests
  - Manufacturing and reprocessing processes similar to the Light Water Reactors (LWR) fuel industrial processes, taking advantage of LWR experience and existing facilities
  - Fuel cycle : well known fabrication process and large experience on reprocessing
  - Scenario : flexible towards Pu management (% and grade), Uranium use and high capabilities for Minor Actinides transmutation



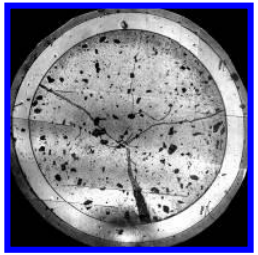
Phénix - CEA

- Metallic fuels
  - High breeding ratios through high fissile density.
  - High thermal conductivity but low melting points which restrict the operating temperatures. Zirconium addition increase the melting point.
  - Limitation may due to fuel clad chemical interaction with low melting eutectics (FCCI). Zirconium liner improves the chemical compatibility.
  - Design optimised to manage the high fuel swelling with low smear density and Na bond.
  - Simplified fabrication without radiotoxic dust risk.



EBR2 - INL

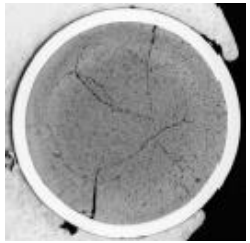
- Carbide fuels



FBTR - IGCAR

  - High breeding ratios through high fissile density.
  - High thermal conductivity and high melting point
  - Design optimised to manage the high fuel swelling with low smear density and Na bond.
  - Carburization of steel
  - R&D needed on fabrication under controlled atmosphere and risk of pyrophoricity
  - Less experience in fabrication and reprocessing than oxide and metal

- Nitride fuels



BOR60 - RIAR

  - High breeding ratios through high fissile density.
  - High thermal conductivity and high melting point
  - Design optimised to manage the high fuel swelling with low smear density and Na bond.
  - Needs for N15 enrichment to avoid C14 production
  - Less experience in fabrication and reprocessing than oxide and metal

SOURCES

- **Bibliography**
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  - Advances in Nuclear Chemistry, M.H.A. Piro, Elsevier 2020.
  - The nuclear fuel of pressurized water reactors and fast neutron reactors, H. Bailly, et al. 1999, Pergamon Press. 660.
  - H. Matzke, Science of Advance LMFBR Fuels. Amsterdam: North-Holland (1986)
  - Dedicated reports or **TECDOC from IAEA** and **Sate Of the Art Reports from OECD/NEA**
  - Publications mainly from ***Journal of Nuclear Materials***, ***Nuclear Engineering and Design*** and ***Nuclear Technology***

- **Courses/school** : FJOH, ISNE, ENEN

- **Conferences** ; FR09, 13, 17, GLOBAL 1993 →2019, IEMPT, NUMAT, ATALANTE

- **Current International activities on oxide and others fuels for advanced reactors :**
  - OECD: expert group on innovative fuel elements (NSC/WPFC/EGIF)
  - AIEA : CRP fuels and materials for fast reactors
  - GIF : Advanced fuel project management board in the SFR, GFR and VHTR systems
  - EUROPEAN PROJECTS : ESFR-SMART, INSPYRE, PUMMA

- **International data bases:**
  - http://therpro.hanyang.ac.kr/search/search_map.jsp
  - https://www.oecd-nea.org/science/taf-id/taf-id-public/
  - https://www.oecd-nea.org/science/wprs/fuel/ifpelst-request.html

Prototypes & industrial SFRs