











# Introduction to the Physics of the Molten Salt Fast Reactor

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For the 'MSFR Team' of LPSC - M. ALLIBERT, M. BROVCHENKO, V. GHETTA, D. HEUER, A. LAUREAU, E. MERLE-LUCOTTE, P. RUBIOLO

With the support of the IN2P3 institute and the PACEN and NEEDS Programs of CNRS, and of the EVOL Euratom FP7 Project

## Liquid fuelled-reactors

#### Which constraints for a liquid fuel?

- Melting temperature not too high
- High boiling temperature
- Low vapor pressure
- Good thermal and hydraulic properties (fuel = coolant)
- Stability under irradiation
- Good solubility of fissile and fertile matters
- No production of radio-isotopes hardly manageable
- Solutions to reprocess/control the fuel salt

Thorium /233U Fuel Cycle

Best candidates = fluoride salt (LiF - 99.995% of  $^{7}$ Li)



#### **Molten Salt Reactors**



Neutronic properties of F not favorable to the U/Pu fuel cycle

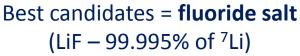
#### **Advantages of a Liquid Fuel**

- ✓ Homogeneity of the fuel (no loading plan)
- ✓ Heat produced directly in the heat transfer fluid
- ✓ Possibility to reconfigure quickly and passively the geometry of the fuel (gravitational draining)
- One configuration optimized for the electricity production managing the criticality
- An other configuration allowing a long term storage with a passive cooling system
- ✓ Possibility to reprocess the fuel without stopping the reactor:
- Better management of the fission products that damage the neutronic and physicochem. properties
- No reactivity reserve (fertile/fissile matter adjusted during reactor operation)

## Liquid fuelled-reactors: MSR

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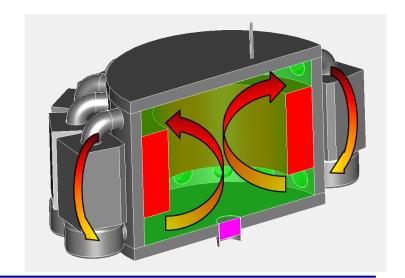
### Thorium /233U Fuel Cycle



Molten Salt Reactor (molten salt = liquid fuel also used as coolant)

Based on the Thorium fuel cycle

With no solid (i.e. moderator) matter in the core ⇒ Fast neutron spectrum



## From MSR to Molten Salt Fast Reactor (MSFR)

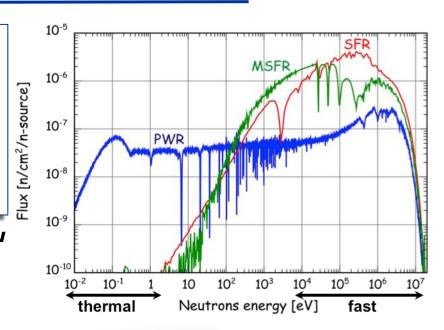
### **Neutronic Optimization of MSR** (Gen4 criteria):

- Safety: negative feedback coefficients
- Sustainability: reduce irradiation damages in the core
- Deployment: good breeding of the fuel + reduced initial fissile inventory

PhD Thesis of L. Mathieu

2008: Definition of an innovative MSR concept based on a fast neutron spectrum, and called MSFR (Molten Salt Fast Reactor) by the GIF Policy Group

- All feedback thermal coefficients negative
- No solid material in the high flux area: reduction of the waste production of irradiated structural elements and less in core maintenance operations
- **▶** Good breeding of the fissile matter thanks to the fast neutron spectrum
- Actinides burning improved thanks to the fast neutron spectrum





#### **R&D** objectives

The renewal and diversification of interests in molten salts have led the MSR provisional SSC to shift the R&D orientations and objectives initially promoted in the original Generation IV Roadmap issued in 2002, in order to encompass in a consistent body the different applications envisioned today for fuel and coolant salts.

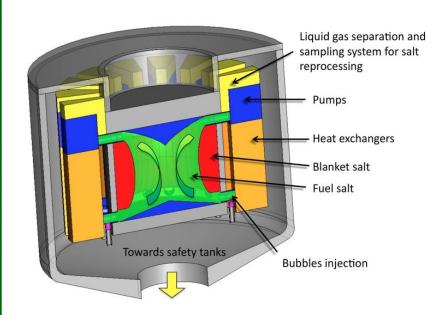
Two baseline concepts are considered which have large commonalities in basic R&D areas, particularly for liquid salt technology and materials behavior (mechanical integrity, corrosion):

- The Molten Salt Fast-neutron Reactor (MSFR) is a long-term alternative to solidfuelled fast neutron reactors offering very negative feedback coefficients and simplified fuel cycle. Its potential has been assessed but specific technological challenges must be addressed and the safety approach has to be established.
- The AHTR is a high temperature reactor with better compactness than the VHTR and passive safety potential for medium to very high unit power.

# The concept of Molten Salt Fast Reactor (MSFR)

Thermal power	3000 MWth
Mean fuel salt temperature	750 °C
Fuel salt temperature rise in the core	100 °C
Fuel molten salt - Initial composition	77.5% LiF and 22.5% [ThF <sub>4</sub> + (Fissile Matter)F <sub>4</sub> ] with Fissile Matter = $^{233}$ U / $^{enriched}$ U / Pu+MA
Fuel salt melting point	565 °C
Fuel salt density	4.1 g/cm <sup>3</sup>
Fuel salt dilation coefficient	8.82 10 <sup>-4</sup> / °C
Fertile blanket salt - Initial composition	LiF-ThF <sub>4</sub> (77.5%-22.5%)
Breeding ratio (steady- state)	1.1
Total feedback coefficient	-5 pcm/K
Core dimensions	Diameter: 2.26 m Height: 2.26 m
Fuel salt volume	18 m³ (½ in the core + ½ in the external circuits)
Blanket salt volume	7.3 m <sup>3</sup>
Total fuel salt cycle	3.9 s

#### Design of the 'reference' MSFR



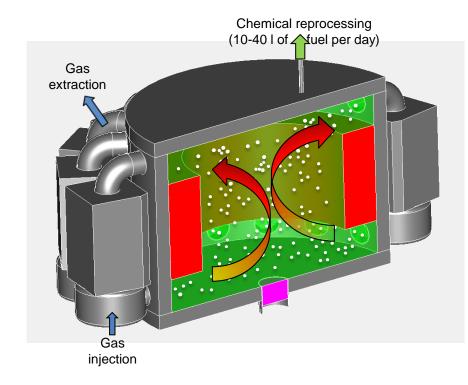
#### **Optimization Criteria:**

Initial fissile matter (233U, Pu, enriched U), salt composition, fissile inventory, reprocessing, waste management, deployment capacities, heat exchanges, structural materials, design...

## MSFR: R&D collaborations

#### 4th Generation reactors => Breeder reactors

Fuel reprocessing mandatory to recover the produced fissile matter – Liquid fuel = reprocessing during reactor operation



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#### 4th Generation reactors => Breeder reactors

Fuel reprocessing mandatory to recover the produced fissile matter – Liquid fuel = reprocessing during reactor operation

Conclusions of the studies: very low impact of the reprocessings (chemical and bubbling) on the neutronic behavior of the MSFR thanks to the fast neutron spectrum = neutronic and chemical (physicochemical properties of the salt) studies driven in parallel

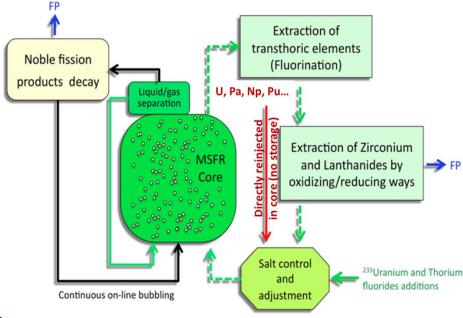
#### PhD Thesis of X. Doligez

Studies requiring multidisciplinary expertise (reactor physics, chemistry, safety, materials, design...)



#### Collaboration at different levels:

- **World:** Generation 4 International Forum
- **► Europe:** Collaborative Project EVOL Euratom/Rosatom + SNETP SRIA Annex
- ➢ <u>National</u>: IN2P3/CNRS and interdisciplinary programs PACEN and NEEDS (CNRS, CEA, IRSN, AREVA, EdF), structuring project 'CLEF' of Grenoble INP



## MSFR and the European project EVOL

European Project "EVOL" Evaluation and Viability Of Liquid fuel fast reactor FP7 (2011-2013): Euratom/Rosatom cooperation

**Objective:** to propose a design of MSFR by end of 2013 given the best system configuration issued from physical, chemical and material studies

- Recommendations for the design of the core and fuel heat exchangers
- Definition of a safety approach dedicated to liquid-fuel reactors Transposition of the defence in depth principle Development of dedicated tools for transient simulations of molten salt reactors
- Determination of the salt composition Determination of Pu solubility in LiF-ThF4 Control of salt potential by introducing Th metal
- Evaluation of the reprocessing efficiency (based on experimental data) FFFER project
- Recommendations for the composition of structural materials around the core



**WP2: Design and Safety** 

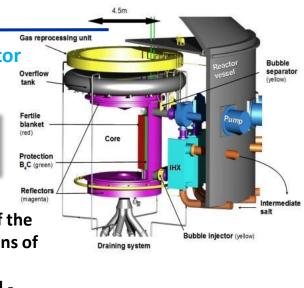
**WP3: Fuel Salt Chemistry and Reprocessing** 

**WP4: Structural Materials** 

12 European Partners: France (CNRS: Coordinateur, Grenoble INP, INOPRO, Aubert&Duval), Pays-Bas (Université Techno. de Delft), Allemagne (ITU, KIT-G, HZDR), Italie (Ecole polytechnique de Turin), Angleterre (Oxford), Hongrie (Univ Techno de Budapest) + 2 observers since 2012 : Politecnico di Milano et Paul Scherrer Institute

+ Coupled to the MARS (Minor Actinides Recycling in Molten Salt) project of ROSATOM (2011-2013)

Partners: RIAR (Dimitrovgrad), KI (Moscow), VNIITF (Snezinsk), IHTE (Ekateriburg), VNIKHT (Moscow) et MUCATEX (Moscow)



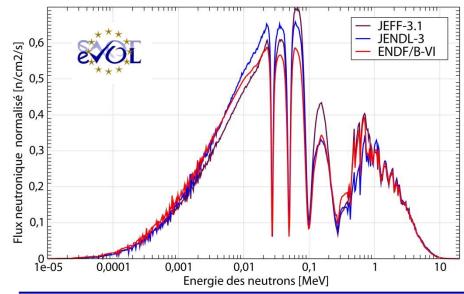


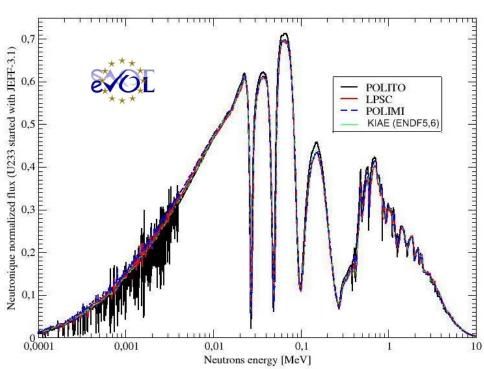
## MSFR optimization: neutronic benchmark (EVOL)

LPSC-IN2P3 calculations performed with a Monte-Carlo neutronic tool (MCNP) coupled to a material evolution code (REM)

Initial Fuel Salt Composition – EVOL Benchmark					
<sup>233</sup> U-start	-started MSFR TRU-started MSFF		MSFR		
Th	<sup>233</sup> U	Th	Actinides		
38 281 kg	4 838 kg	30 619 kg	Pu	11 079 kg	
				5.628 %mol	
19.985 %mol	2.515 %mol	16.068 %mol	Np	789 kg	
				0.405 %mol	
			Am	677 kg	
				0.341 %mol	
			Cm	116 kg	
				0.058 %mol	

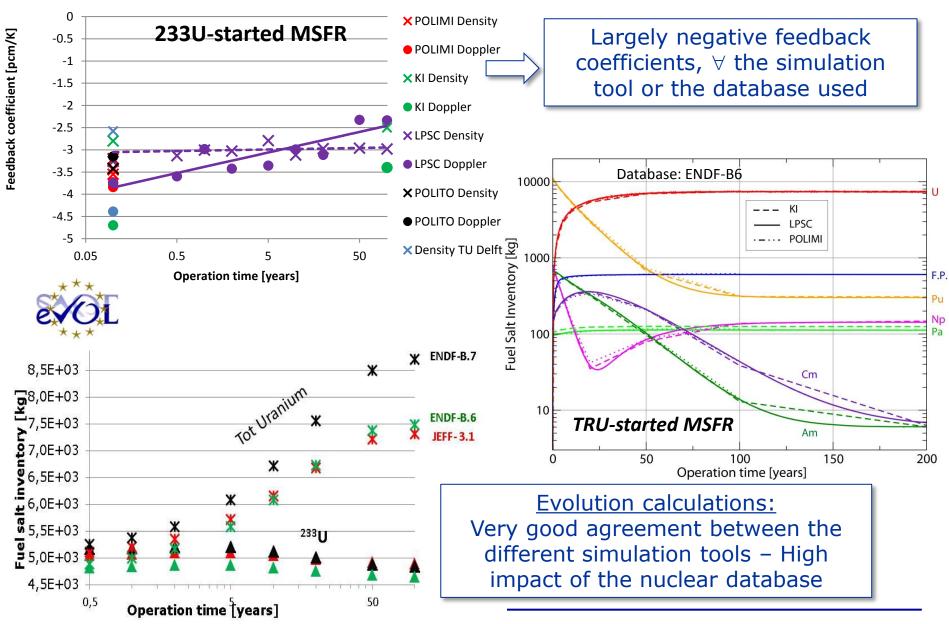
#### PhD Thesis of M. Brovchenko





Static calculations (BOL here):
Good agreement between the different simulation tools – High impact of the nuclear database

# MSFR optimization: neutronic benchmark (EVOL)



## MSFR optimization: initial fissile matter

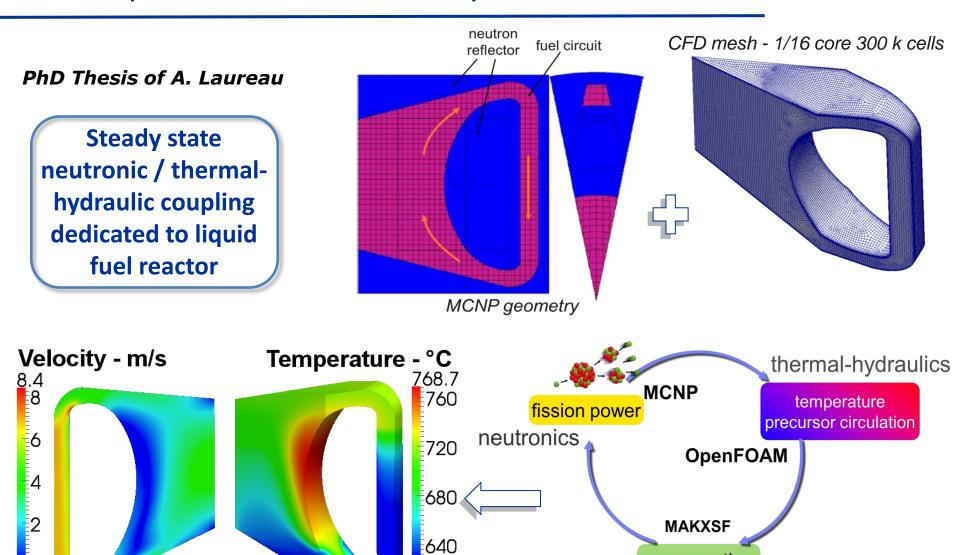
#### Which initial fissile load to start a MSFR?

- Start directly <sup>233</sup>U produced in Gen3+ or Gen4 (included MSFR) reactors
- Start directly with enriched U: U enrichment < 20% (prolif. Issues)</li>
- Start with the Pu of current LWRs mixed with other TRU elements: solubility limit of valence-III elements in LiF
- Mix of these solutions: Thorium as fertile matter +
  - ➤ <sup>233</sup>U + TRU produced in LWRs
  - ➤ MOx-Th in Gen3+ / other Gen4
  - Uranium enriched (e.g. 13%) + TRU currently produced

[kg per GWe]	<sup>233</sup> U started MSFR	TRU (Pu UOx)	Enriched U (13%) + TRU started MSFR	Th Pu-MOx started MSFR
Th 232	25 553	20 396	10 135	18 301
Pa 231				20
U 232				1
U 233	3 260			2 308
U 234				317
U 235			1 735	45
U 236				13
U 238			11 758	
Np 237		531	335	54
Pu 238		229	144	315
Pu 239		3 902	2 464	1 390
Pu 240		1 835	1 159	2 643
Pu 241		917	579	297
Pu 242		577	364	1 389
Am 241		291	184	1 423
Am 243		164	104	354
Cm 244		69	44	54
Cm 245		6	4	

Thorium Energy Conference 2013 (ThEC13) – CERN, Gel

## MSFR optimization: thermal-hydraulic studies



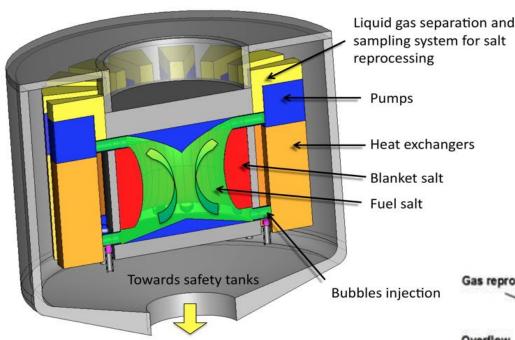
624.8

cross section

nuclear database

**ENDF-B7** 

# Molten Salt Fast Reactor (MSFR): fuel circuit



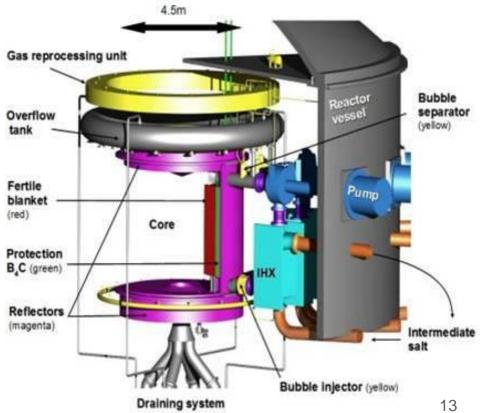
## Core (active area):

No inside structure

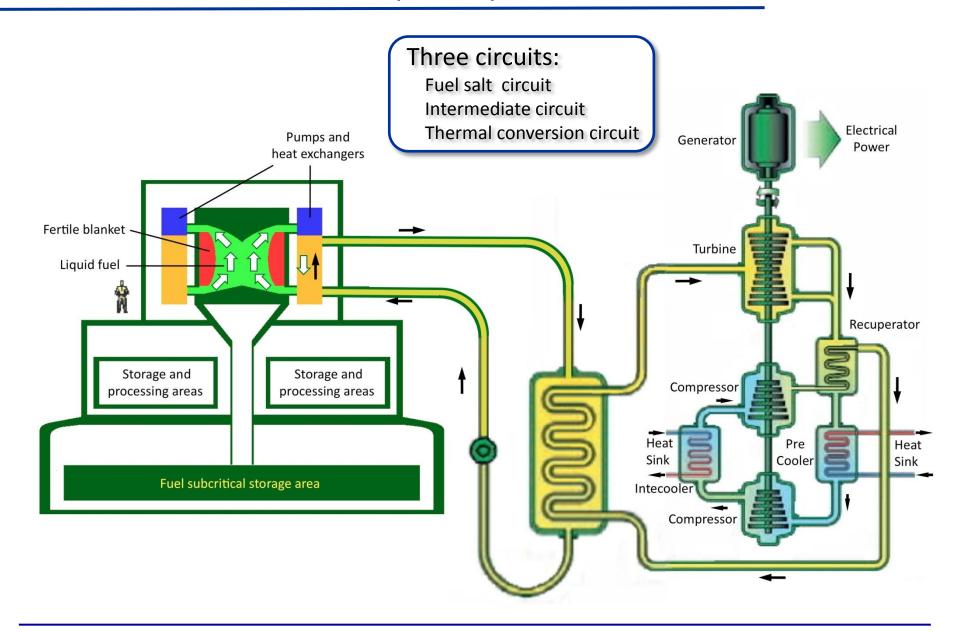
Outside structure: Upper and lower Reflectors, Fertile Blanket Wall

## + 16 external recirculation loops:

- Pipes (cold and hot region)
- Bubble Separator
- Pump
- Heat Exchanger
- Bubble Injection



# Molten Salt Fast Reactor (MSFR)



## **MSFR** and Safety Evaluation

## Design aspects impacting the MSFR safety analysis

## Liquid fuel

- ✓ Molten fuel salt acts as reactor fuel and coolant
- ✓ Relative uniform fuel irradiation
- ✓ A significant part of the fissile inventory is outside the core
- ✓ Fuel reprocessing and loading during reactor operation

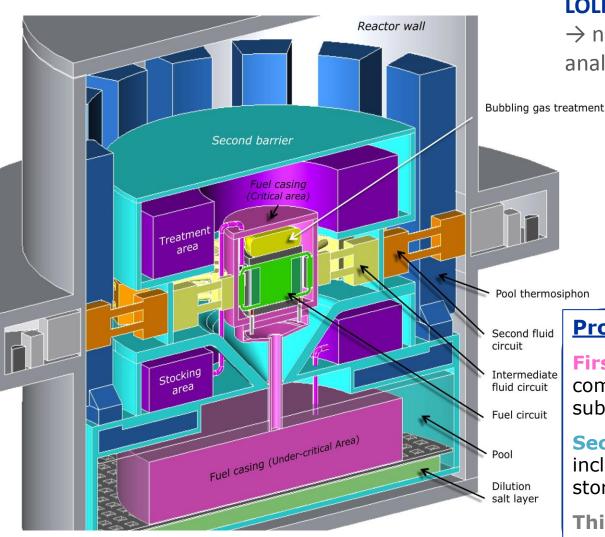
#### No control rods in the core

- ✓ Reactivity is controlled by the heat transfer rate in the HX + fuel salt feedback coefficients, continuous fissile loading, and by the geometry of the fuel salt mass
- ✓ No requirement for controlling the neutron flux shape (no DNB, uniform fuel irradiation, etc.)

### Fuel salt draining

- ✓ Cold shutdown is obtained by draining the molten salt from the fuel circuit
- ✓ Changing the fuel geometry allows for adequate shutdown margin and cooling.
- ✓ Fuel draining can be done passively or by operator action

## MSFR and Safety Evaluation



#### **LOLF accident (Loss of Liquid Fuel)**

→ no tools available for quantitative analysis but qualitatively:

- Fuel circuit: complex structure, multiple connections
- Potential leakage: collectors connected to draining tank



#### **Proposed Confinement barriers:**

First barrier: fuel envelop, composed of two areas: critical and sub-critical areas

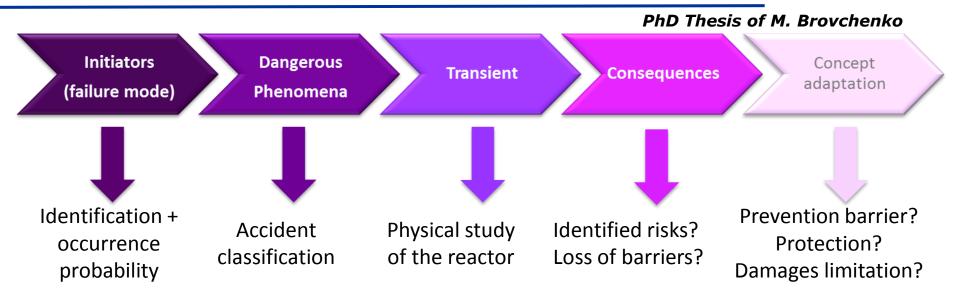
**Second barrier**: reactor vessel, also including the reprocessing and storage units

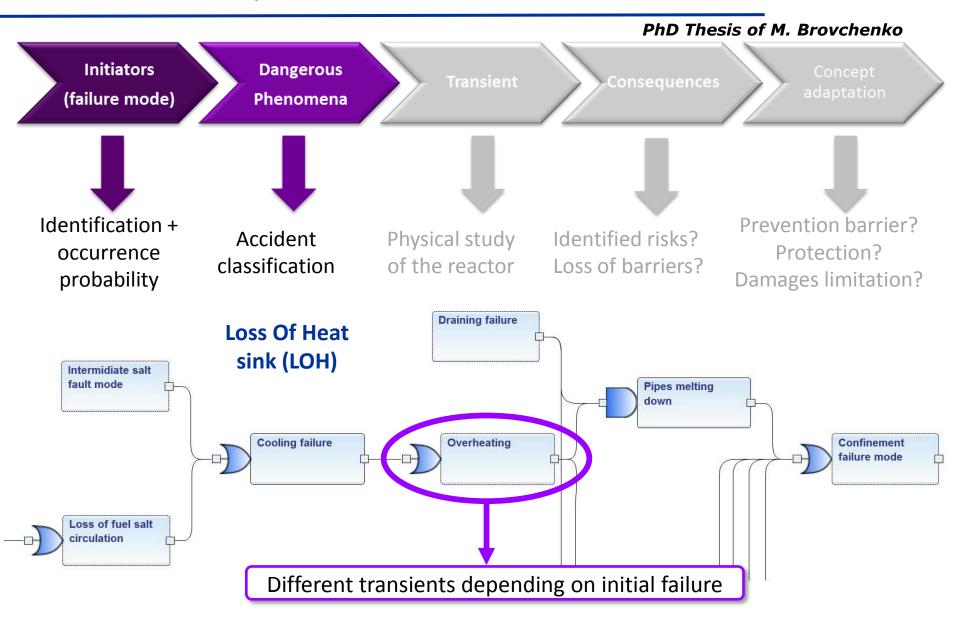
Third barrier: reactor wall, corresponding to the reactor building

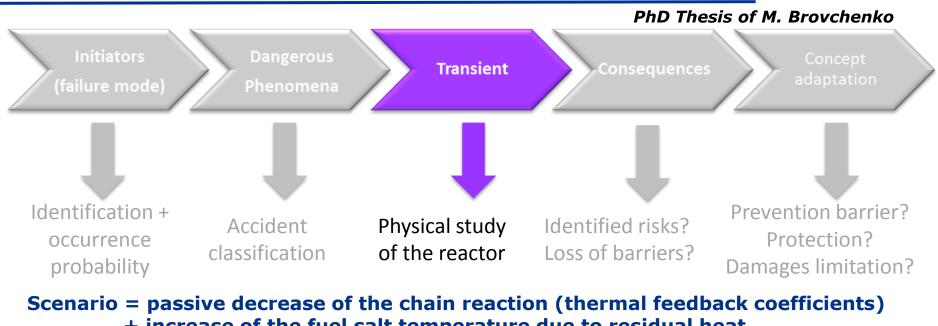
## MSFR and Safety Evaluation

## **Safety analysis: objectives**

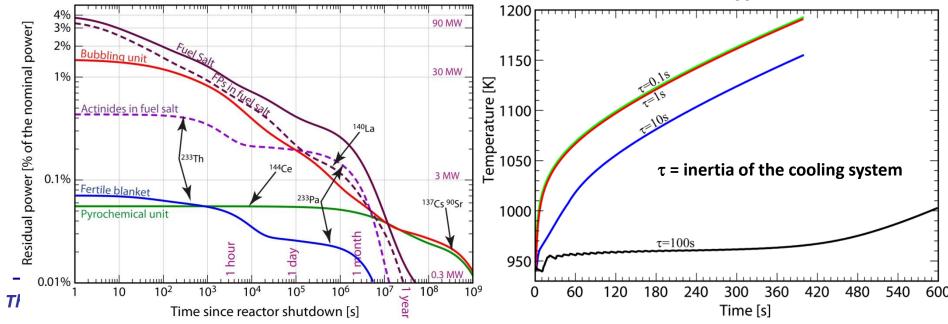
- Develop a safety approach dedicated to MSFR
  - **Based on current safety principles** e.g. defense-in-depth, multiple barriers, the 3 safety functions (reactivity control, fuel cooling, confinement) etc. but adapted to the MSFR.
  - Integrate both **deterministic and probabilistic** approaches
  - Specific approach dedicated to **severe accidents**:
    - Fuel liquid during normal operation
    - Fuel solubility in water (draining tanks)
    - Source term evaluation
- Build a reactor risk analysis model
  - Identify the **initiators and high risk scenarios** that require detailed transient analysis
  - Evaluate the risk due to the **residual heat and the radioactive inventory** in the whole system, including the reprocessing units (chemical and )
  - Evaluate some potential design solutions (barriers)
  - Allow reactor designer to estimate impact of design changes (design by safety)

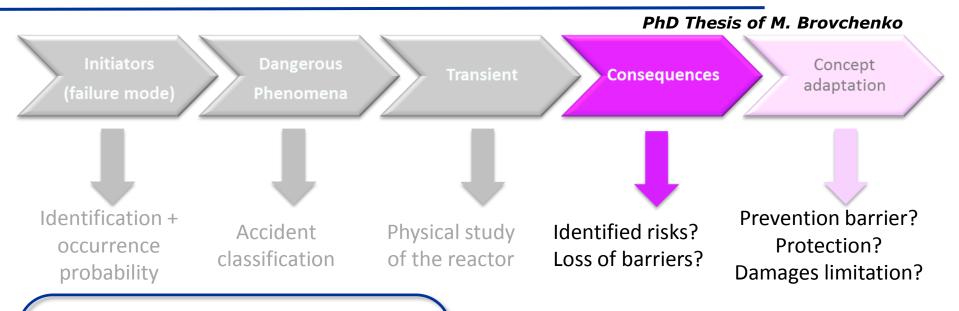






# + increase of the fuel salt temperature due to residual heat





## **Risks identified:**

- Continuous heating due to the residual power (physics)
- Increase of temperature : impact of the pump inertia (technology)

#### **Quantitatively:** Risk = Probability x Severity

Accident probabilities and severity difficult to quantify at the current preliminary design stage

#### **Protection:**

- Draining of the fuel salt
- Thermal protection on the walls?

'Design by Safety' approach

## **Demonstration and Demonstrator of MSFR**

#### **Sizing of the facilities:**

<u>Small size:</u> ~1liter - chemistry and corrosion – off-line processing Pyrochemistry: basic chemical data, processing, monitoring

Medium size: ~100 liters – hydrodynamics, noble FP extraction, heat exchanges Process analysis, modeling, technology tests

Full size experiment: ~1 m³ salt / loop – validation at loop scale

Validation of technology integration and hydrodynamics models

### 3 levels of radio protection:

- ✓ Inactive simulant salt ⇒ Standard laboratory
   Hydrodynamics, material, measurements, model validation
- ✓ Low activity level (Th, depleted U) ⇒ Standard lab + radio protect Pyrochemistry, corrosion, chemical monitoring
- ✓ High activity level (enriched U, 233 U, Pu, MA) 

  ⇒ Nuclear facility

  Fuel salt processing: Pyrochemistry, , Actinides recycling

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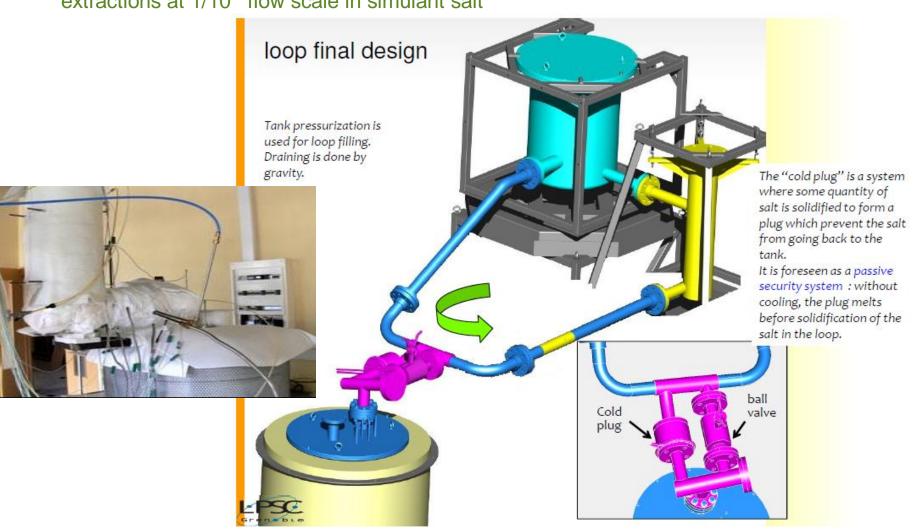
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## Demonstration and Demonstrator of MSFR: the FFFER facility

## The Forced Fluoride Flow Experiment

Reproduces the gases and particles extractions at 1/10<sup>th</sup> flow scale in simulant salt



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## Power Demonstrator of the MSFR

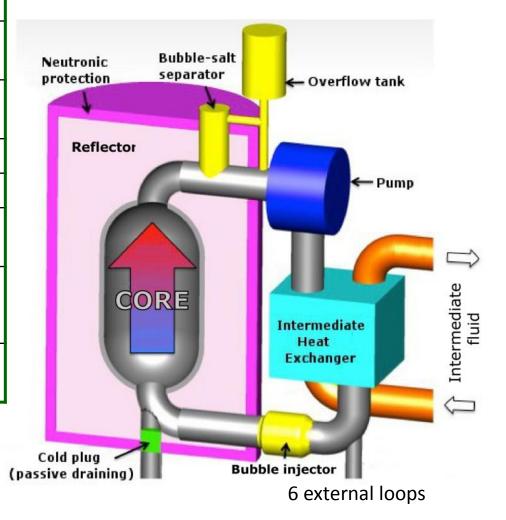
Thermal power	100 MWth
Mean fuel salt temperature	725 °C
Fuel salt temperature rise in the core	30 °C
Fuel Molten salt initial composition	75% LiF-ThF <sub>4</sub> - <sup>233</sup> UF <sub>4</sub> or LiF- ThF <sub>4</sub> -( <sup>enriched</sup> U+MOx-Th)F <sub>3</sub>
Fuel salt melting point	565 °C
Fuel salt density	4.1 g/cm <sup>3</sup>
Core dimensions	Diameter: 1.112 m Height: 1.112 m
Fuel Salt Volume	1.8 m <sup>3</sup> 1.08 in core 0.72 in external circuits
Total fuel salt cycle in the fuel circuit	3.5 s

Demonstrator characteristics

representative of the MSFR

#### From the power reactor to the demonstrator:

Power / 30 and Volume / 10

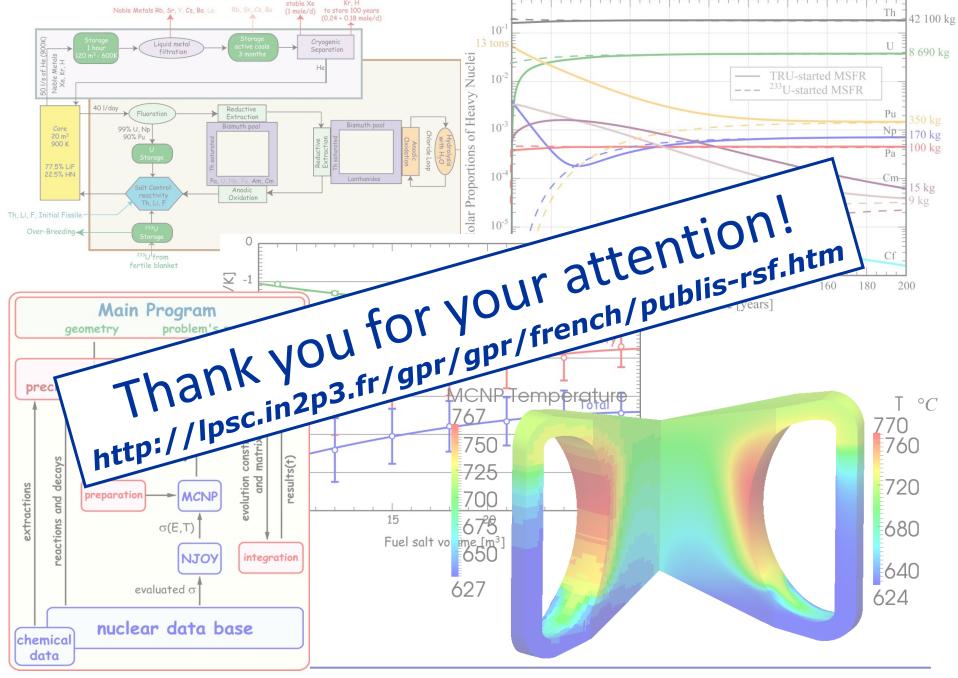


## **MSFR**: Conclusions and Perspectives

**Summary:** Definition of an innovative Molten Salt configuration with a Fast Neutron Spectrum, based firstly on reactor physics studies and including now more largely system developments (chemistry, thermal-hydraulics, materials, safety, design...)

#### **Perspectives**

- ⇒ Where?
- National programs: CNRS (IN2P3...) and multidisciplinary program NEEDS Collaborations with IRSN (and EdF/AREVA?) + Structuring project CLEF of Grenoble INP
- European project EVOL (FP7) with Rosatom: finished end 2013 Next project in Horizon 2020?
- International: MSR MoU (GIF) to be signed by ROSATOM Other collaborations (China, Japan, USA...)?
  - ⇒ Optimization of the system and symbiotic safety/design studies
- Multi-physics and multi-scale coupling tool for a global simulation of the system
- Design of the reactor, draining and processing systems (including materials, components...)
- Risk analysis and safety approach dedicated to MSFR
- Define the demonstration steps and experimental facilities



# MSFR: choice of the liquid fluid

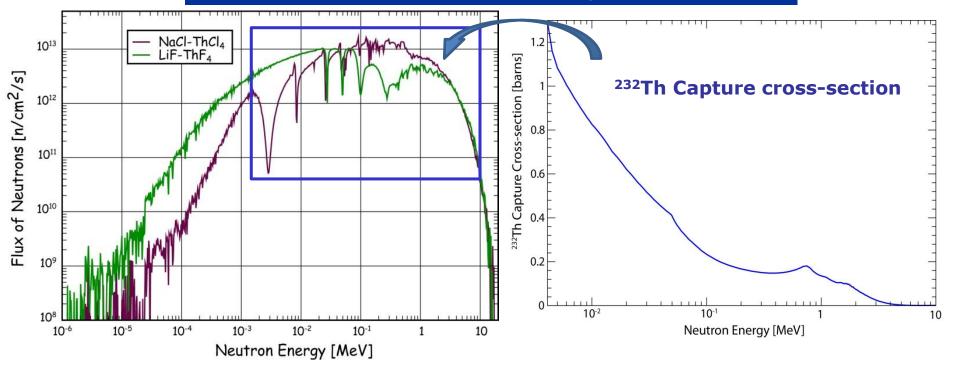
Element produced	Problem	Fluoride Salt	Chloride Salt
<sup>36</sup> Cl produced via <sup>35</sup> Cl(n,γ) <sup>36</sup> Cl and <sup>37</sup> Cl(n,2n) <sup>36</sup> Cl	Radioactivity - T <sub>1/2</sub> = 301000y		10 moles / y (373 g/year)
$^3$ H produced via $^6$ Li(n, $lpha$ ) t and $^6$ Li(n,t) $lpha$	Radioactivity - T <sub>1/2</sub> = 12 years	55 moles / y (166 g/y)	
Sulphur produced via $^{37}$ Cl(n, $\alpha$ ) $^{34}$ P( $\beta$ -[12.34s]) $^{34}$ S and $^{35}$ Cl(n, $\alpha$ ) $^{32}$ P( $\beta$ -[14.262 days]) $^{32}$ S	Corrosion (located in the grain boundaries)		10 moles / year
Oxygen produced via $^{19}$ F(n, $lpha$ ) $^{16}$ O	Corrosion (surface of metals)	88.6 moles/year	
Tellurium produced via fissions and extracted by the on-line bubbling	Corrosion (cf. Sulphur)	200 moles/year	200 moles/year

Combination of both neutronic and chemical considerations

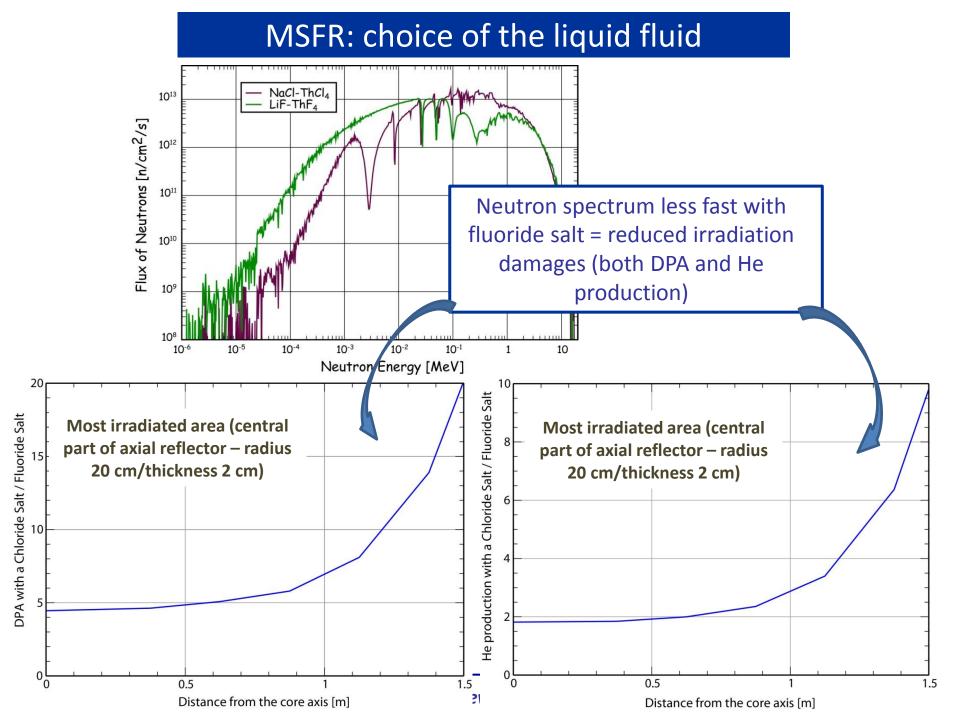


MISER based on a molten Life fuel salt

## MSFR: choice of the liquid fluid



Parameter	Fluoride Salt	Chloride Salt
Thorium capture cross-section in core (barn)	0.61	0.315
Thorium amount in core (kg)	42 340	47 160
Thorium capture rate in core (mole/day)	11.03	8.48
Thorium capture cross-section in blanket (barn)	0.91	0.48
Thorium amount in the blanket (kg)	25 930	36 400
Thorium capture rate in the blanket (mole/day)	1.37	2.86
<sup>233</sup> U initial inventory (kg)	5720	6867
Neutrons per fission v in core	2.50	2.51
<sup>233</sup> U capture cross-section in core (barn)	0.495	0.273
<sup>233</sup> U fission cross-section in core (barn)	4.17	2.76
Capture/fission ratio $\alpha$ (spectrum-dependent)	0.119	0.099
Thorium Energy ( Total breeding ratio	1.126	1.040



"Fuel Salt Loop" = Includes all the systems in contact with the fuel salt during normal operation

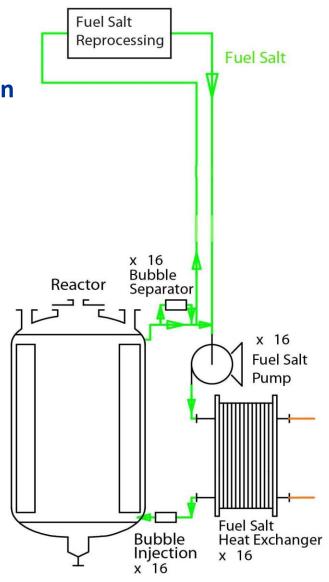
#### Core:

No inside structure

Outside structure: Upper and lower Reflectors, Fertile Blanket Wall

#### + 16 external modules:

- Pipes (cold and hot region)
- Bubble Separator
- Pump
- Heat Exchanger
- Bubble Injection

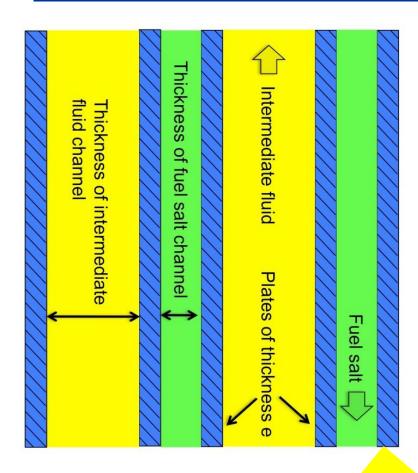




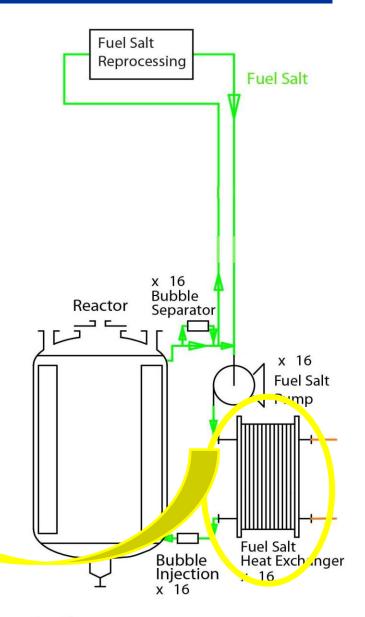








Two kind of intermediate fluid considered in this study: liquid metal or fluoride salt











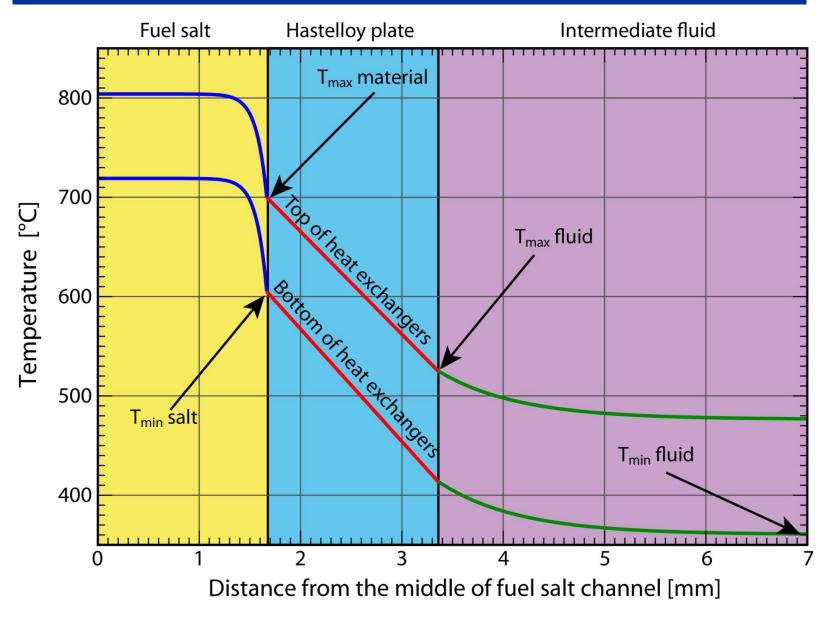
Constrained Parameter	Limiting value (P <sub>0i</sub> )	Acceptable deviation (σ <sub>i</sub> )
Minimum thickness of the fuel salt channel	2.5 mm	0.05 mm
Minimum thickness of the plate	1.75 mm	0.035 mm
Maximum speed of the fuel salt	3.5 m/s	0.07 m/s
Maximum speed of the intermediate fluid (liquid lead)	1.75 m/s	0.035 m/s
Maximum speed of the intermediate fluid (salt)	5.5 m/s	0.11 m/s
Maximum temperature of the materials	700 °C	1 °C
Minimum margin to solidification of the fuel salt	50 °C	1°C
Minimum margin to solidification of the intermediate fluid	40 °C	1 °C

# Each set of values of the variable parameters evaluated with the quality function: $\prod exp\left(\frac{P_i-P_{0i}}{\sigma_i}\right)$

#### Variables of the study:

- ✓ the diameter of the pipes
- ✓ the thickness of the plates
- ✓ the gap between the plates on the intermediate fluid side (or "thickness of the intermediate fluid channel")
- ✓ the fuel salt temperature at core entrance
- ✓ the fuel salt temperature increase within the core
- ✓ the temperature increase of the intermediate fluid in the heat exchangers
- ✓ the mean temperature difference between the two fluids within the heat exchangers

Evaluated parameter	Pb	FLiNaK	NaF-NaBF <sub>4</sub>
Diameter of the fuel salt pipes [mm]	301	283	303
Diameter of the intermediate fluid pipes [mm]	897	507	470
Thickness of the plates [mm]	1.61	1.51	1.65
Fuel salt temperature at core entrance [°C]	754	698	704
Fuel salt temperature increase in the core [°C]	89	106	98
Intermediate fluid temperature increase within the heat exchangers [°C]	99	41	66
Mean temperature difference between the two fluids in the heat exchangers [°C]	382	242	280
Intermediate fluid temperature at the heat exch. outlet [°C]	466	530	506
Thickness of the fuel salt channel [mm]	3.38	2.17	2.37
Thickness of the intermediate fluid channel [mm]	29.8	4.49	4.38
Fuel salt speed in the pipes [m/s]	3.92	3.97	3.73
Fuel salt speed in the heat exchangers [m/s]	3.85	2.36	2.91
Intermediate fluid speed in the pipes [m/s]	1.94	6.00	5.67
Intermediate fluid speed in the heat exchangers [m/s]	1.92	5.54	5.75
Maximum temperature of the intermediate fluid [°C]	523	622	595
Maximum temperature of the materials [°C]	701	701	699
Margin to the solidification of the fuel salt [°C]	43.7	54.7	46.7
Margin to the solidification of the intermediate fluid [°C]	39.6	34.5	56.2
Pressure loss of the fuel salt in the heat exchangers [bar]	2.56	2.03	2.56
Pressure loss of the fuel salt in the pipes [bar]	0.99	1.02	0.90
Pressure loss of the intermediate fluid in the heat exch. [bar]	0.09	2.09	1.66
h Pressure loss of the intermediate fluid in the pipes [bar]	0.32	0.71	0.57



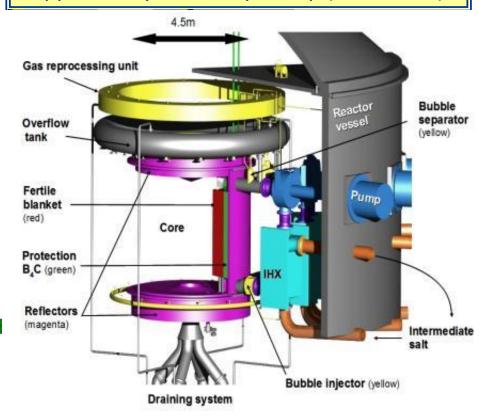
# The concept of Molten Salt Fast Reactor

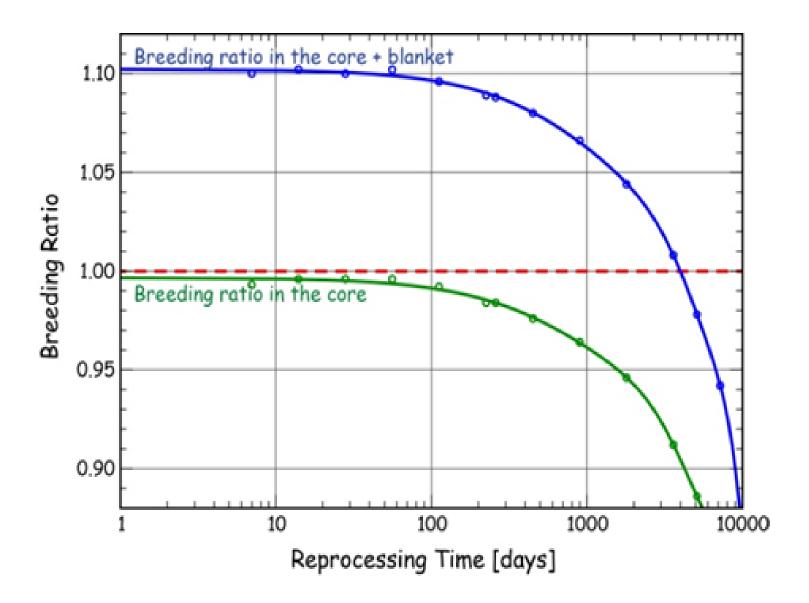
### Design of the reference MSFR

- Initial Salt: 77.5%LiF 2.5% <sup>233</sup>UF<sub>3</sub> ThF<sub>4</sub>
- <sup>233</sup>U initial inventory per GW<sub>él</sub>: 3260 kg
- <sup>233</sup>U production (breeder reactor): 95 kg/year
- Feedback Coefficient: -5 pcm/K
- Fuel Salt Temperature: 750 °C
- Produced power: 3 GW<sub>th</sub> (~1.5 GW<sub>el</sub>)
- Core Internal Diameter = Core Height = 2.3 m
- Fuel Salt Volume: 18 m<sup>3</sup>
  - 1/2 in the active zone (core + plenums)
  - 1/2 in the external circuit (heat exchangers, pipes, pumps)
- Thickness of Fertile Blanket: 50 cm
- Volume of Fertile Blanket: 7.7 m<sup>3</sup>
- Initial Fertile Salt: 77.5%LiF 22.5%ThF<sub>4</sub>
- Core reprocessing: 10 to 40 l of fuel salt cleaned per day (on-site batch reprocessing for lanthanides extraction) + on-line He bubbling in the core

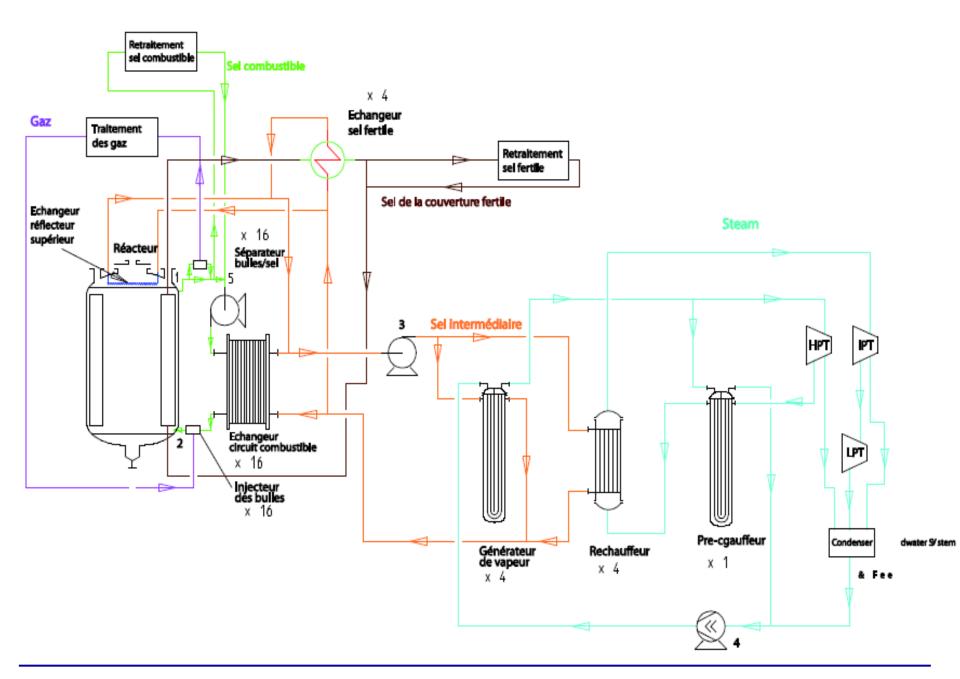


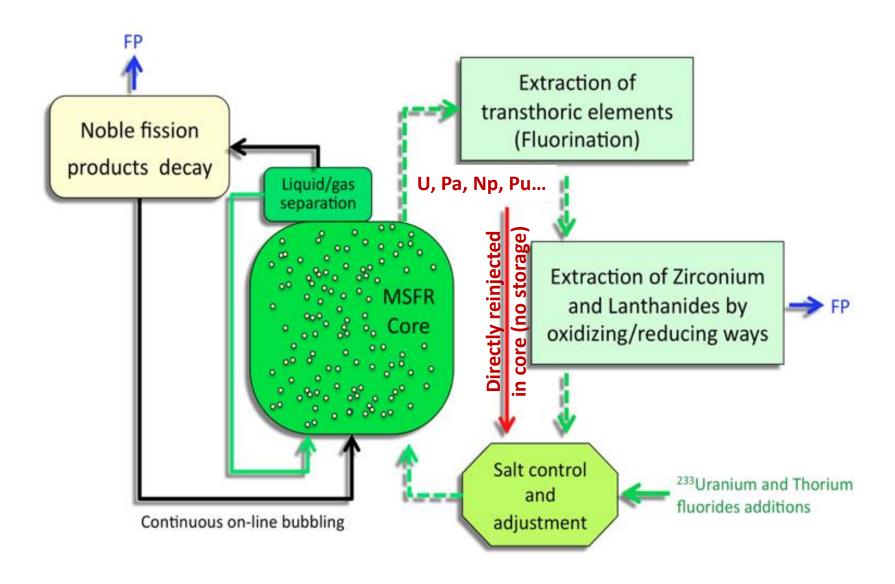
MSFR concept selected for further studies by the GIF "MSR Steering Committee" – Choice approved by the Policy Group (since 2008)





Initial Fuel Salt Composition – EVOL Benchmark								
Initial I	uel Salt Com	position – EVO	L Ben	chmark				
<sup>233</sup> U-start	ed MSFR	TRU-s	tartec	MSFR				
Th	<sup>233</sup> U	Th		Actinides				
38 281 kg	4 838 kg	30 619 kg	Pu	11 079 kg				
				5.628 %mol				
19.985 %mol	2.515 %mol	16.068 %mol	Np	789 kg				
				0.405 %mol				
			Am	677 kg				
				0.341 %mol				
			Cm	116 kg				
				0.058 %mol				





# Liquid fuelled-reactors: why "molten salt reactors"?

#### Which constraints for a liquid fuel?

- Melting temperature not too high
- High boiling temperature
- Low vapor pressure
- Good thermal and hydraulic properties (fuel = coolant)
- Stability under irradiation
- Good solubility of fissile and fertile matters
- No production of radio-isotopes hardly manageable
- Solutions to reprocess/control the fuel salt

Lithium fluorides fulfill all constraints

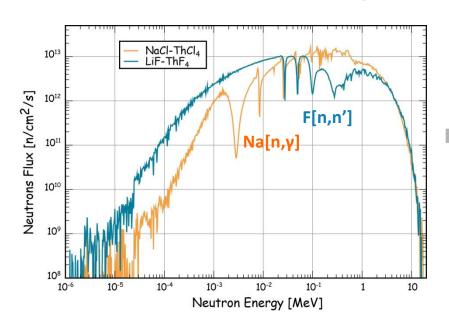


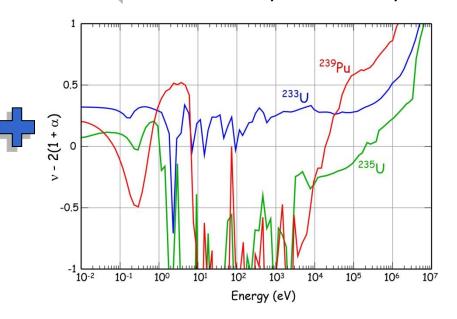
**Molten Salt Reactors** 



Neutronic cross-sections of fluorine versus neutron economy in the fuel cycle

#### Thorium /233U Fuel Cycle





# Molten Salt Reactor (MSR): Historical studies

# Historical studies of MSR: Oak Ridge Nat. Lab. - USA

• 1954 : Aircraft Reactor Experiment (ARE)

Operated during 1000 hours

Power = 2.5 MWth



**Experimental Reactor** 

Power: 7.4 MWth

Temperature: 650°C

U enriched 30% (1966 - 1968)

<sup>233</sup>U (1968 – 1969) - <sup>239</sup>Pu (1969)

No Thorium inside

• 1970 - 1976: Molten Salt Breeder Reactor (MSBR)

Never built

Power: 2500 MWth

Thermal neutron spectrum





# Future of nuclear reactors: 4th Generation Systems

#### **Generation 4 International Forum: Criteria for Future Nuclear Reactors**

#### Sustainable development

- ➤ Availability
  - Long term availability of the system
  - Resources availability → Reactors at least breeder
- ➤ Minimization of the waste production
  - Recycling of Actinides + Minimizing the MA production
  - Minimizing the Industrial Wastes (structural elements and processes)
- ➤ Deployment capacities
  - Minimizing the Initial Fissile Inventory versus breeding
  - Availability of the Initial Fissile Matter

### **Optimal Safety and Reliability**

- ➤ Reduction of major accident/incident's initiators
- ➤ Risks and consequences of core damages limited
  - No inflammable matters in the core, no high pressure
  - Minimized reactivity margins
  - All negative safety coefficients

<u>Proliferation Resistance and Physical Protection</u>

**Economic Competitiveness** 

⇒ Development of innovative MSR concepts to fulfill these criteria

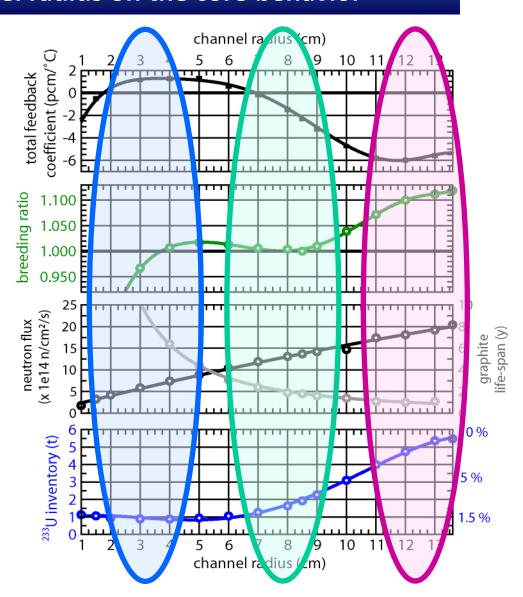


# **Historical MSR Studies at CNRS**

#### Influence of the channel radius on the core behavior

# Three types of configuration:

- thermal (r = 3-6 cm)
- epithermal (r = 6-10 cm)
- fast (r > 10 cm)



# **Historical MSR Studies at CNRS**

Influence of the channel radius on the core behavior

# Thermal spectrum configurations

- positive feedback coefficient
- iso-breeder
- quite long graphite life-span
- low <sup>233</sup>U initial inventory

PhD thesis of Ludovic MATHIEU

# **Epithermal spectrum configurations**

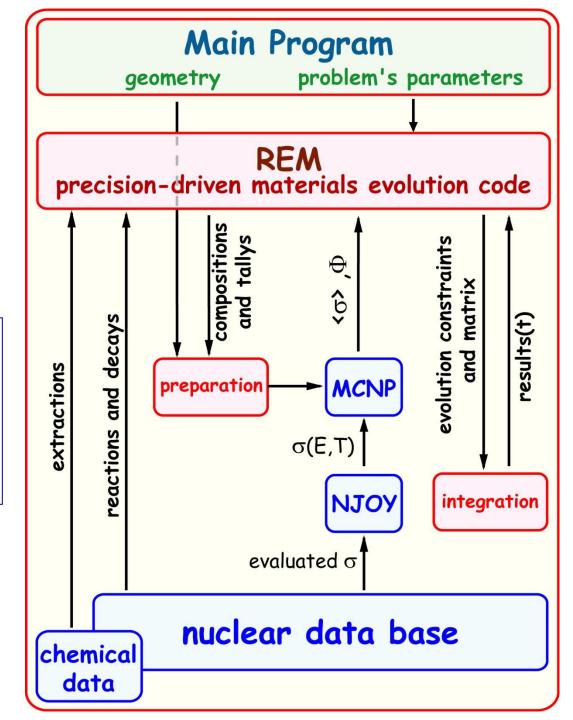
- quite negative feedback coefficient
- iso-breeder
- very short graphite life-span
- quite low <sup>233</sup>U initial inventory

# Fast spectrum configurations (no moderator)

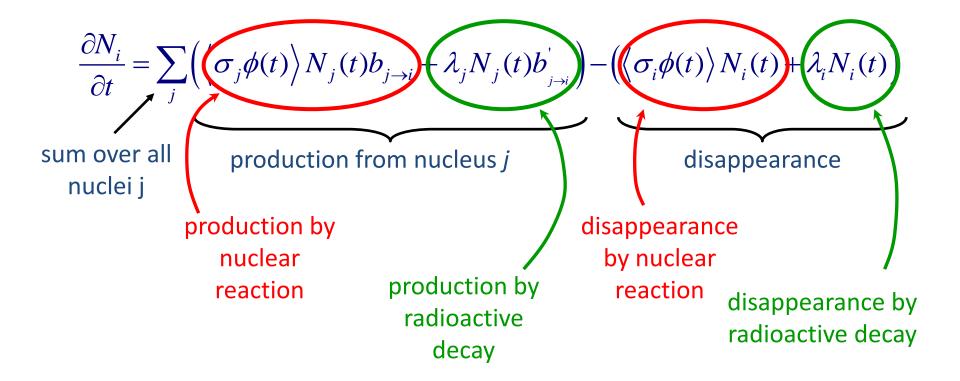
- very negative feedback coefficients
- very good breeding ratio
- no problem of graphite life-span
- large <sup>233</sup>U initial inventory

Tools for the
Simulation of Reactor
Evolution:
Details of the program

code REM for materials
evolution with the
probabilistic code MCNP for
neutronic calculations



# Tools for the Simulation of Reactor Evolution: Integration Module: Bateman Equation for nucleus i



Molten Salt Reactors: addition of a feeding term, equal to the number of nuclei added per time unit for each element (flow)

Reprocessing: new terms  $-\lambda_i^{extr.}N_i \quad \text{with} \quad \lambda_i^{extr.} = \frac{1}{T_i^{reprocess.}}$  Efficiency linked to the nucleus extraction probability

#### **Fission Products Extraction: Motivations**

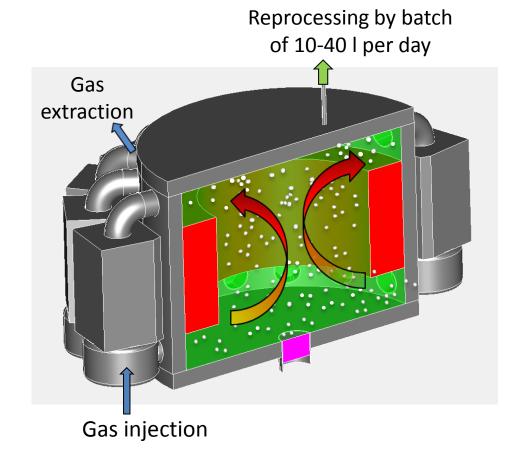
- ✓ Control physicochemical properties of the salt (control deposit, erosion and corrosion phenomena's)
- √ Keep good neutronic properties

### **Physical Separation (in the core)**

- ➤ Gas Reprocessing Unit through bubbling extraction
- Extract Kr, Xe, He and particles in suspension

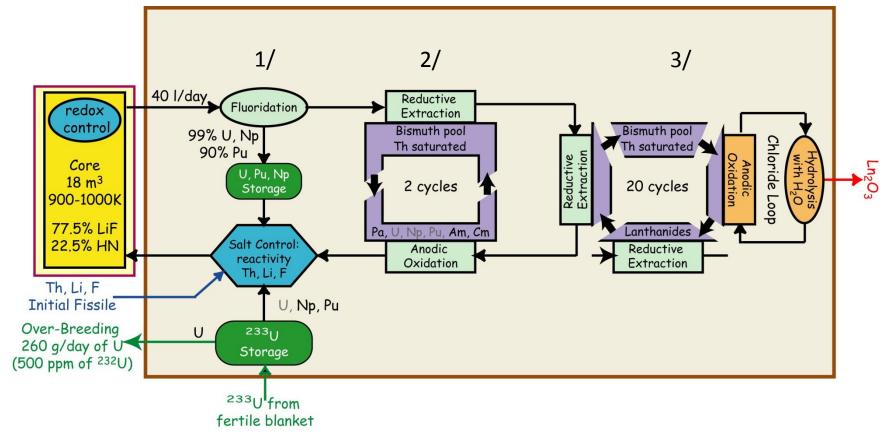
### **Chemical Separation (by batch)**

- ➤ Pyrochemical Reprocessing Unit
- ➤ Located on-site, but outside the reactor vessel



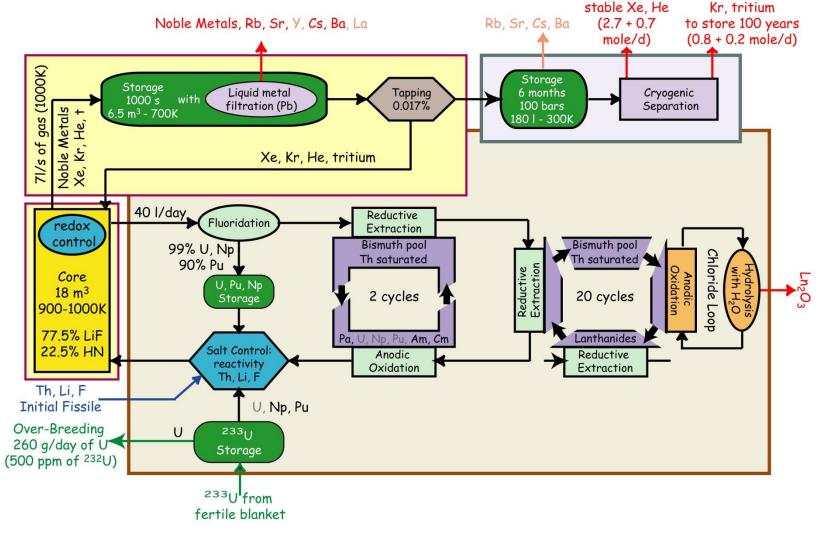
#### **On-site Chemical Reprocessing Unit**

- 1/ Salt Control + Fluorination to extract U, Np, Pu + few FPs Expected efficiency of 99% for U/Np and 90% for Pu Extracted elements re-injected in core
- 2/ Reductive extraction to remove actinides (except Th) from the salt MA re-injected by anodic oxidation in the salt at the core entrance
- 3/ Second reductive extraction to remove all the elements other than the solvent lanthanides transferred to a chloride salt before being precipitated



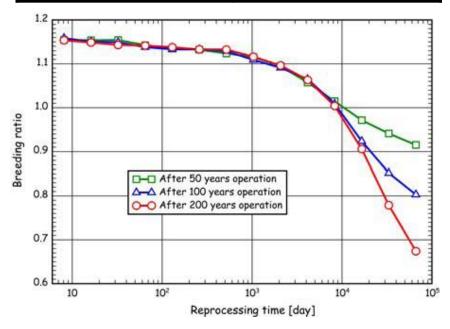
#### Noble gazes bubbling in the core (within the fuel salt loop)

To remove all insoluble fission products (mostly noble metals) and rare gases, helium bubbles are voluntary injected in the flowing liquid salt (bottom of the core)  $\rightarrow$  Separation salt / bubbles  $\rightarrow$  Treatment on liquid metal and then cryogenic separation (out of core)

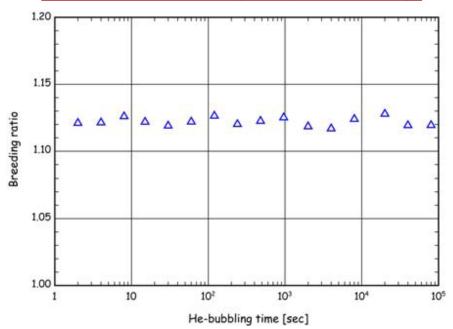


# **Batch reprocessing:**

Element	Absorption (per fission neutron)		
Heavy Nuclei	0.9		
Alkalines	< 10 <sup>-4</sup>		
Metals	0.0014		
Lanthanides	0.006		
Total FPs	0.0075		



### On-line (bubbling) reprocessing:



#### **Fast neutron spectrum**

- ⇒ very low capture cross-sections
  - ⇒ low impact of the FP extraction on neutronics
- ⇒ Parallel studies of chemical and neutronic issues possible

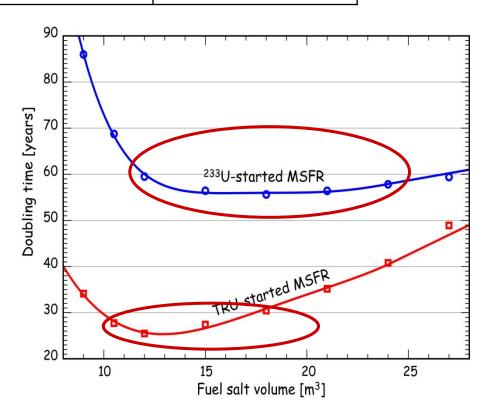
# MSFR: Design and Fissile Inventory Optimization

Fuel salt volume / specific power	t(100 dpa)	t(100 ppm He)	t(-1 at% of W)
12 m <sup>3</sup> - 500 W/cm <sup>3</sup>	85 years	2.2 years	4.7 years
18 m <sup>3</sup> – 330 W/cm <sup>3</sup>	133 years	3.2 years	7.3 years
27 m <sup>3</sup> - 220 W/cm <sup>3</sup>	211 years	5.5 years	10.9 years



**Optimization = Medium Fuel Salt Volumes** 





# MSFR: Design and Fissile Inventory Optimization

Reactor Design and Fissile Inventory Optimization = Specific Power Optimization

#### 2 parameters:

- The produced power
- The fuel salt volume and the core geometry

Liquid fuel and no solid matter inside the core ⇒ possibility to reach specific power much higher than in a solid fuel

### 3 limiting factors:

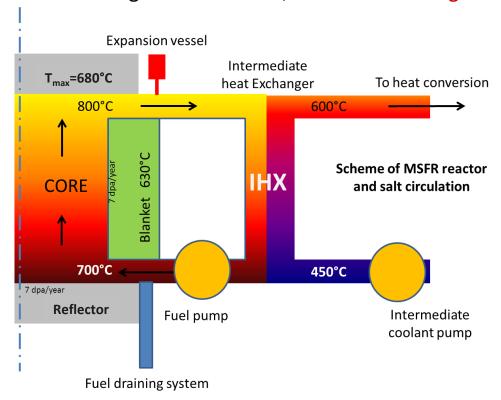
- The capacities of the heat exchangers in terms of heat extraction and the associated pressure drops (pumps)  $\rightarrow$  large fuel salt volume and small specific power
- ullet The neutronic irradiation damages to the structural materials which modify their physicochemical properties. Three effects: displacements per atom, production of Helium gas, transmutation of Tungsten in Osmium o large fuel salt volume and small specific power
- The neutronic characteristics of the reactor in terms of burning efficiencies  $\rightarrow$  small fuel salt volume and large specific power and of deployment capacities, i.e. breeding ratio (=  $^{233}$ U production) versus fissile inventory  $\rightarrow$  optimum near  $15m^3$  and  $400W/cm^3$ 
  - ⇒ Reference MSFR configuration with 18m³ et 330 W/cm³ corresponding to an initial fissile inventory of 3.5 tons per GWe

# 4<sup>th</sup> Generation International Forum and MSFR: Availability

## MSFR Availability: structural materials (Ni-based alloys) resistance

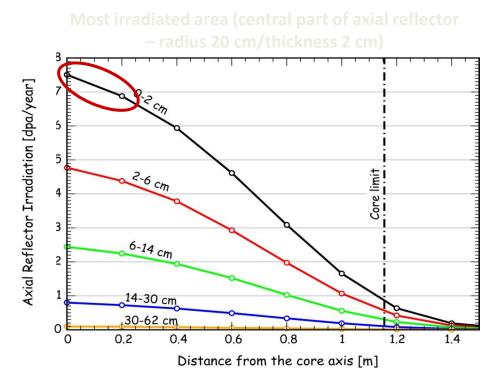
Ni	W	Cr	Мо	Fe	Ti	С	Mn	Si	Al	В	Р	S
79.432	9.976	8.014	0.736	0.632	0.295	0.294	0.257	0.252	0.052	0.033	0.023	0.004

Neutronic irradiation damages to the structural materials (modify their physicochemical properties) = displacements per atom, production of Helium gas, transmutation of Tungsten in Osmium, activation – At high temperatures



# 4<sup>th</sup> Generation International Forum and MSFR: Availability

<u>Displacements per atom</u>: represent the number of times one atom is displaced for a given neutron flux



+ Effects due to fissions occurring near the material wall - damages on the first tens µm

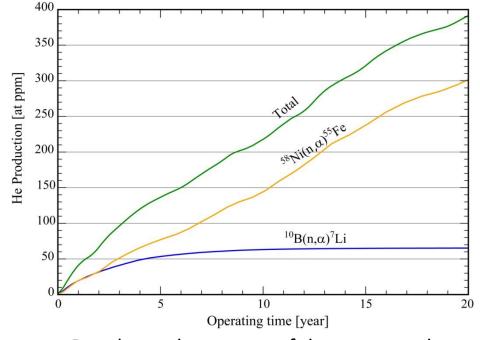
# Main activated elements in structural materials

	T <sub>1/2</sub> [years]	[At/cm³]	Decay mode
<sup>59</sup> Ni	76000	2.97 10 <sup>20</sup>	EC
<sup>63</sup> Ni	99	3.56 10 <sup>19</sup>	β <sup>-</sup> 67 keV
<sup>99</sup> Tc	211300	1.26 10 <sup>19</sup>	β <sup>-</sup> 294 keV
<sup>93</sup> Mo	3012	2.85 10 <sup>18</sup>	EC +88% 31 keV
<sup>93</sup> Nb	16	1.75 10 <sup>15</sup>	IT 31 keV
<sup>3</sup> H	12	1.23 10 <sup>15</sup>	β <sup>-</sup> 19 keV

# 4<sup>th</sup> Generation International Forum and MSFR: Availability

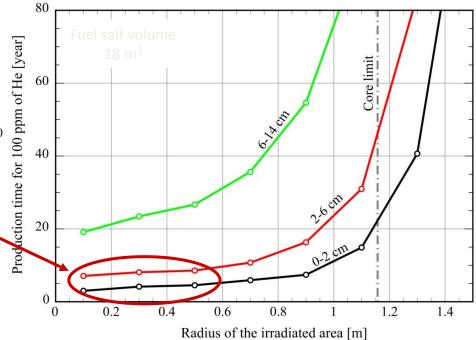
#### Helium production in the structural materials

Ni	w	Cr	Мо	Fe	Ti	С	Mn	Si	Al	В	Р	S
79.432	9.976	8.014	0.736	0.632	0.295	0.294	0.257	0.252	0.052	0.033	0.023	0.004



⇒ Regular replacements of these area to be planned (first 10cm only) or enriched Ni (lower <sup>58</sup>Ni content) or addition of a thin layer of another material (SiC?) to protect the surface of these reflectors

Main contribution to Helium production in the most irradiated area (radius 20 cm /thickness 2 cm) for a fuel salt volume of 18 m<sup>3</sup> due to <sup>58</sup>Ni



# 4th Generation International Forum and MSFR: Availability

#### Transmutation of the Tungsten contained in the alloy into Rhenium and Osmium

Ni	W	Cr	Мо	Fe	Ti	С	Mn	Si	Al	В	Р	S
79.432	9.976	8.014	0.736	0.632	0.295	0.294	0.257	0.252	0.052	0.033	0.023	0.004

<sup>192</sup>P† 0.782%

<sup>191</sup>Ir

193P+

50 y

192**I**r

73.827 d

19105

15.4 d

194P+

32.967%

193Ir

62.7%

<sup>192</sup>Os

40.93%

Transmutation Cycle of W in Re and Os (neutronic captures + decays):

37.3% <sup>189</sup>Os 186Os <sup>187</sup>Os 188Os 190Os 13.29% 16.21% 26.36% 1.59% <sup>185</sup>Re <sup>187</sup>Re 186Re. 188Re. 37.4% 3.72 d 62.6% 17 h 183W 184W/ 185W/ 186W/ 187W 14.31% 30.64% 75 d 28.43% 23.72 h

W, Re and Os contents of the most irradiated area for a fuel salt volume of 18 m<sup>3</sup>:

• Value of the acceptable limit?

182 Ta

114.43 d

<sup>182</sup>W

26.5%

<sup>181</sup>Ta

100%

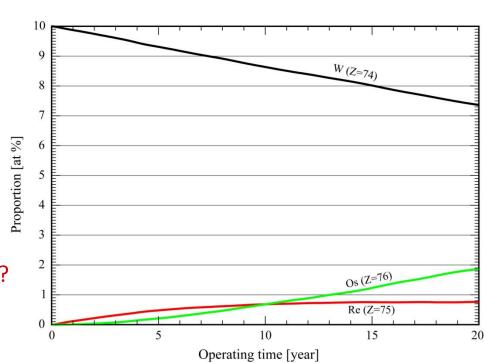
180W

0.12%

181W/

121 d

• Impact on the structural materials resistance?



196P+

197P+

25.242% 19.8915 h

195P+

33.832%

194Tr

10.53 d

193W/

30.11 h

# 4th Generation International Forum and MSFR: Availability

# MSFR Availability: structural materials (Ni-based alloys) resistance

Ni	W	Cr	Мо	Fe	Ti	С	Mn	Si	Al	В	Р	S
79.432	9.976	8.014	0.736	0.632	0.295	0.294	0.257	0.252	0.052	0.033	0.023	0.004

Neutronic irradiation damages to the structural materials (modify their physicochemical properties) = displacements per atom, production of Helium gas, transmutation of Tungsten in Osmium, activation

Structural elements: layers	Displacements per atom	He production	Tungsten transmutation
0-2.5 cm	6.8 dpa/year	12 ppm / year	0.11 at% /year
2.5-7.5 cm	3.5 dpa/year	6 ppm / year	0.07 at% /year

To be experimentally studied: He production (maximal acceptable amount, diffusion effects?) + Effects on the long-term resistance of structural materials due to W transmutation + Effects of high temperature on structural materials

#### **Conclusions:**

- Irradiation damages low + Limits unknown
- Irradiation damages limited to the first 10 cm (replaced 3-4 times or use a thin layer of SiC for example as thermal protection)
- Materials not under large mechanical stress

# Generation4 and Th fuel cycle

Uranium cycle partially used in currently operating reactors (cf MOX fuel)

- + used in Phenix / Superphenix
- + studied in mainly Gen4 reactors

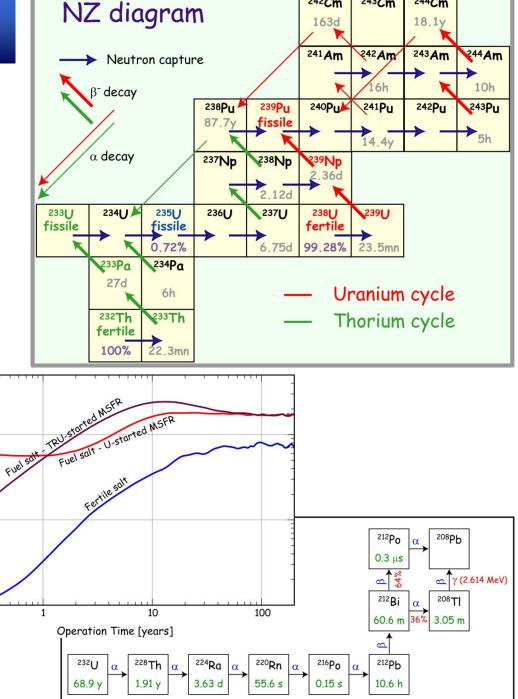
Thorium/ $^{233}$ U fuel cycle = only alternative to U/Pu fuel cycle

Fraction of 232U/U total [ppm]

1000

#### Thorium fuel cycle presents 2 essential advantages:

- Lower production of transuranic elements (TRU)
- High proliferation resistance thanks to the decay of <sup>232</sup>U (2.6 MeV gamma - activity of 1g of <sup>232</sup>U at equilibrium = 270 GBq) mixed with <sup>233</sup>U in the core + blanket

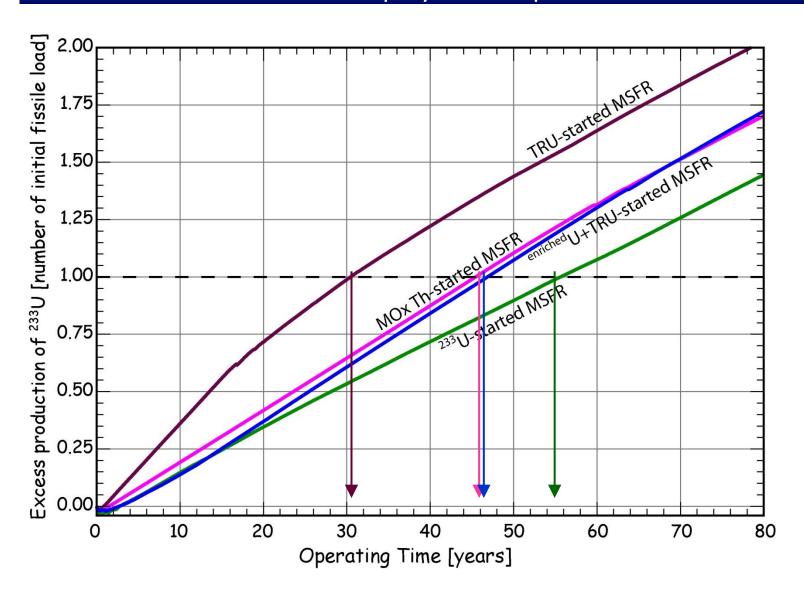


242Cm

243Cm

244Cm

# 4<sup>th</sup> Generation International Forum and MSFR: Starting Modes and Deployment Capacities



# 4<sup>th</sup> Generation International Forum and MSFR: Starting Modes and Deployment Capacities

Deployment scenario at a French scale with a linear doubling of the installed nuclear power between 2020 and 2100 with these assumptions:

- Current PWRs stopped after 45 years of operation
  - > 10% using MOX fuel (corresponding Minor Actinides vitrified)
- EPR fleet: deployed from 2014
  - > From 2040: some of these EPRs loaded with MOX fuel and Thorium
- MSFR fleet deployed in 2070, using the output of MoX-Th irradiated in these EPRs
- As soon as possible (when <sup>233</sup>U available): MSFRs started with a mix of <sup>233</sup>U-PuUoX or <sup>233</sup>U-PuMoX (cf [<sup>233</sup>U+TRU]-started MSFR configurations)
- First half of the XXII<sup>th</sup> century: decision to stop the fission based electricity production (replaced by a novel technology)
  - ➤ Introduction of "incinerator MSFRs" to further reduce the heavy nuclei inventories discharged after the final shutdown of the MSFR fleet

# Deployment scenarios: reduction of the final HN inventory

"Incinerator MSR" identical to MSFR except for the fuel salt composition + suppression of the fertile blanket

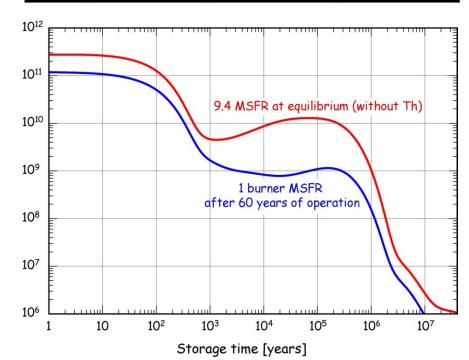
# Fuel salt: FLiNaK with 46.5% <sup>7</sup>LiF, 11.5% NaF, 41.7% KF, (HN)F<sub>4</sub>

- Melting point correctly low even with small HN proportion (no Th) in the salt
- Neutron spectrum not too thermalized

#### **Incinerator operation:**

- Initial HN load to reach criticality: 685 kg of transTh from MSFR
- Fueled with transTh from MSFR to maintain reactivity
   Shutdown after 60 years of operation: HNg
- Shutdown after 60 years of operation: HNg burning equivalent to 9.4 MSFR
   inventories

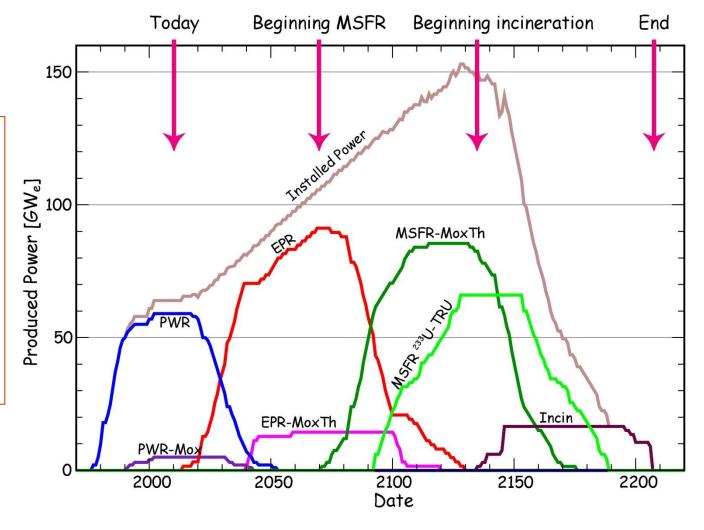
[kg]	9.4 MSFR (input)	Inventory at 60 yrs	Burning efficiency
U	72 751	6 407	11.5
Np	1 381	506	2.8
Pu	2 768	1 530	1.8
Am	72	39	1.8
Cm	33	64	0.5
HN	77 005	8 550	9.1



# 4th Generation International Forum and MSFR: Deployment scenarios of the Th fuel cycle with MSFRs

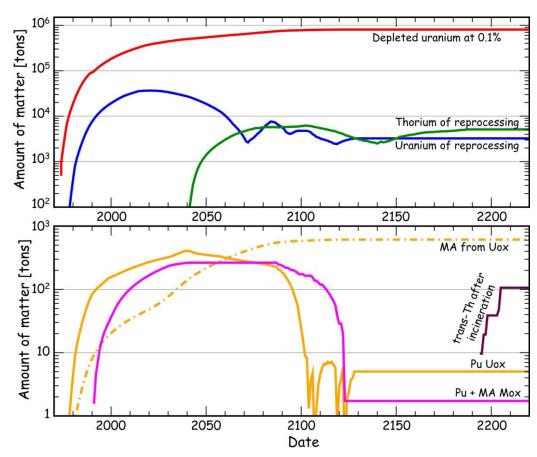
Total power produced = 138 000 TWh among which 72 300 TWh by the MSFR fleet

Very good deployment capacities Transition to the Thorium fuel cycle achieved
+ Close the current fuel cycle (reduce the stockpiles of produced transuranic elements)



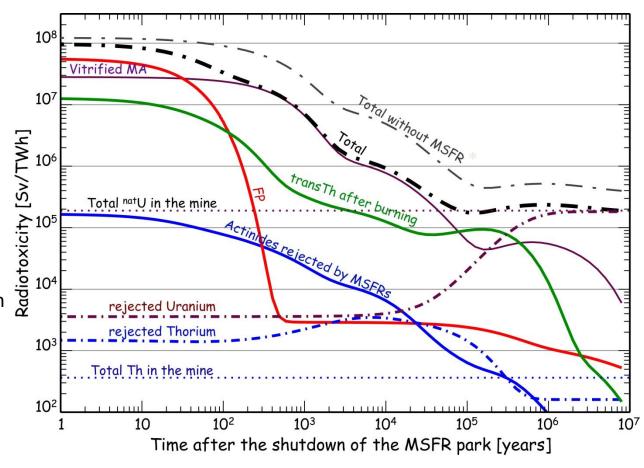
# 4th Generation International Forum and MSFR: Deployment scenarios of the Th fuel cycle with MSFRs

- Stockpiles of uranium from reprocessing largely reduced
- Stockpiles of Pu-Uox, Pu-Mox and AM-Mox totally burned in MSFR ⇒ remains only MA extracted from Uox fuel when using Pu-Mox in PWRs and EPRs
- After incinerator MSFRs: only 100 tons of transthorian elements remaining
- -Around 18 000 t of actinides used for fission (138 000 TWh
  - 11 700 t from natural U
  - 6 300 t from Th
- Natural resources needed for this nuclear deployment:
  - 821 400 t of natural U
  - 11 600 t of Th



# 4th Generation International Forum and MSFR: Deployment scenarios of the Th fuel cycle with MSFRs

- ⇒ Scenario optimized but without MSFR and the Th fuel cycle: radiotoxicity 3 to 5 times higher between 1000 and 100 000 years
- Long term radiotoxicity dominated by the vitrified MA from Uox fuel mixed with the FPs (Gen2 and Gen3 reactors)
- Very long term radio-toxicity (after 300 000 years) dominated by the rejected uranium (depleted + reproc.) see long life decay products of <sup>238</sup>U (as <sup>230</sup>Th and <sup>234</sup>U)
- Radiotoxicity of the transthorian elements from the MSFR fleet (final inventories) lower than the extracted natural U after 3 000 years

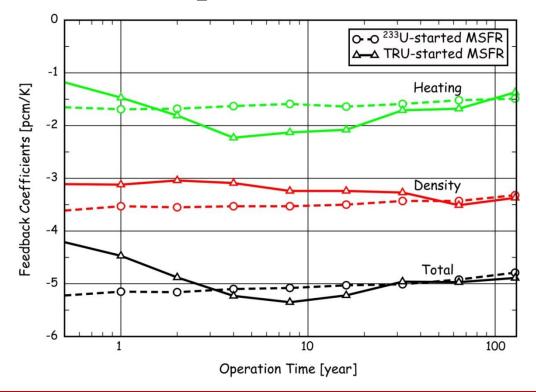


# 4<sup>th</sup> Generation International Forum and MSFR Control of the chain reaction: Neutronic safety parameters

#### 3- Safety parameters: Feedback coefficients

dk/dT = Variation of the multiplication factor (dk) with the core temperature (dT)Reactor intrinsically safe if <math>dk/dT < 0 (if T  $\nearrow$  then k  $\searrow$ )

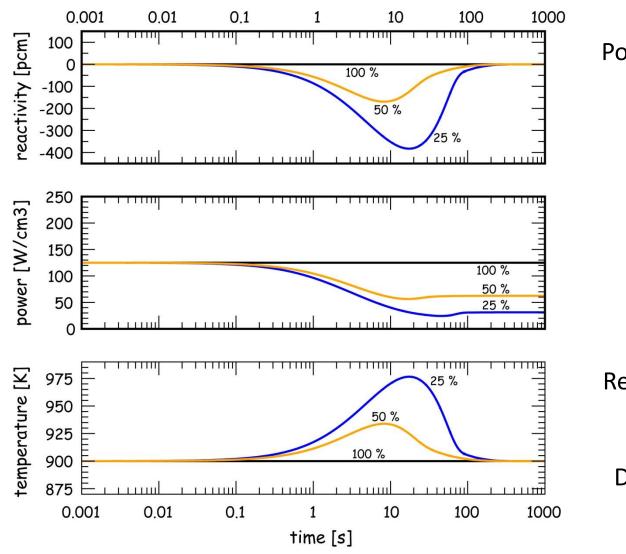
$$\left(\frac{dk}{dT}\right)_{Total} = \left[\frac{dk}{dT}\right]_{Salt Heating} + \left(\frac{dk}{dT}\right)_{Salt Density} + correlations\right] < 0$$



- ⇒ dk/dT largely < 0 for all MSFR configurations and equal to -5 pcm/K for the reference configuration</p>
- + Salt density coefficient (equivalent to void coefficient) < 0 for all configurations too

⇒ MSFR: Only Gen4 system being both breeder and with all negative safety coefficients

# 4<sup>th</sup> Generation International Forum and MSFR Control of the chain reaction & Power demand regulation



Power demand decrease



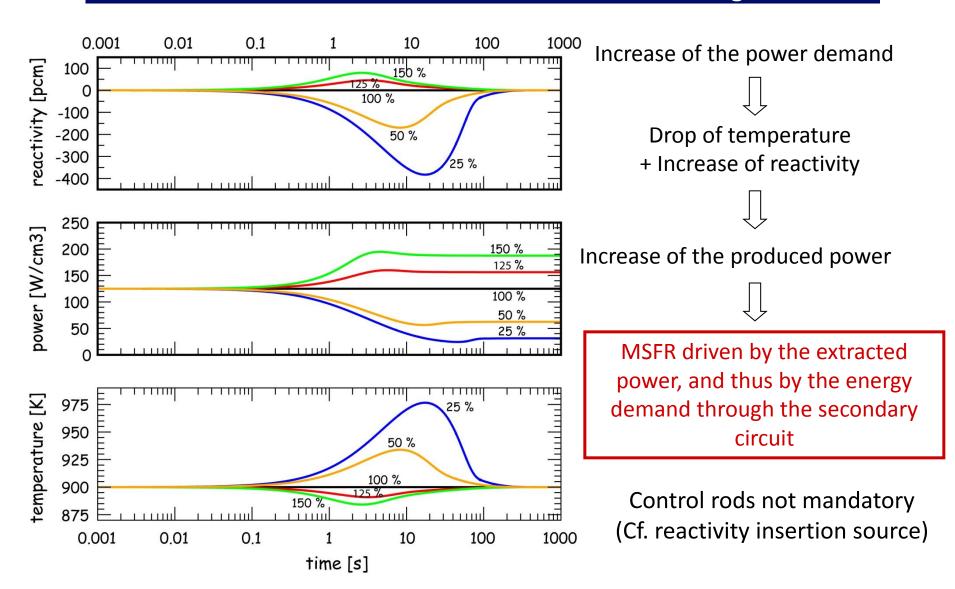
Rise of temperature + Drop of reactivity



Return to equilibrium at the nominal temperature

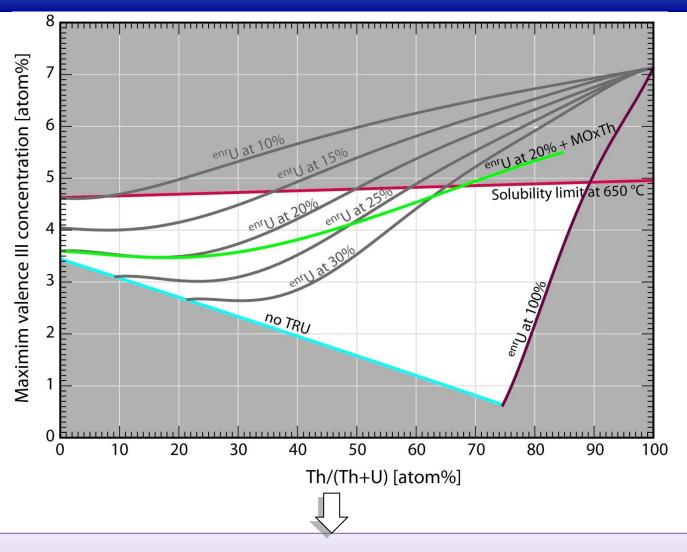
Decrease of the produced power

# 4<sup>th</sup> Generation International Forum and MSFR Control of the chain reaction & Power demand regulation



Prof. E. Merle-Lucotte, INSTN/ENEN Gen4 Seminar, Sept 2012

#### Power Demonstrator of the MSFR: initial fissile load



- √ enriched U mixed with transuranic elements possible with U enrichment of 15% 20%
- ✓ Uranium enriched at 20% mixed with irradiated MOx-Th with a ratio of Th/(Th+U) = 20 to 65%

#### From Power Demonstrator of the MSFR to SMR

	No radial blanket and H/D=1	No radial blanket and H/D=1
Power [MW <sub>th</sub> ]	100	200
Initial <sup>233</sup> U load [kg]	654	654
Fuel reprocessing of 1I/day		
Feeding in <sup>233</sup> U [kg/an]	11.38	23.38
Breeding ratio	-29.83%	-30.64%
Total <sup>233</sup> U needed [kg]	1013.87	1388.37

Around 650kg of <sup>233</sup>U to start

**Under-breeder reactor** 

Fuel reprocessing of 4I/day		
Feeding in <sup>233</sup> U [kg/an]	11.20	22.58
Breeding ratio	-29.37%	-29.59%
Total <sup>233</sup> U needed [kg]	1001.86	1353.13

Low impact of the chemical reprocessing rate (not mandatory for the demonstrator)

# From Power Demonstrator of the MSFR to SMR

	No radial blanket and H/D=1	No radial blanket and H/D=1	Radial blanket and H/D=1	Radial blanket and H/D=1
Power [MW <sub>th</sub> ]	100	200	100	200
Initial <sup>233</sup> U load [kg]	654	654	667	667
Fuel reprocessing of 1I/day				
Feeding in <sup>233</sup> U [kg/an]	11.38	23.38	1.72	4.70
Breeding ratio	-29.83%	-30.64%	-4.52%	-6.16%
Total <sup>233</sup> U needed [kg]	1013.87	1388.37	738.83	835.16
Breeding ratio (radial + axial fertile blankets)			1.81%	-0.04%
Fuel reprocessing of 4I/day				
Feeding in <sup>233</sup> U [kg/an]	11.20	22.58	1. 48	3.58
Breeding ratio	-29.37%	-29.59%	-3.88%	-4.69%
Total <sup>233</sup> U needed [kg]	1001.86	1353.13	722.50	794.21
Breeding ratio (radial + axial fertile blankets)			2.49%	1.54%

Addition of axial + radial fertile blankets ⇒ small modular breeder MSFR

### From Power Demonstrator of the MSFR to SMR

	No radial blanket and H/D=1	No radial blanket and H/D=1	Radial blanket and H/D=1	Radial blanket and H/D=1	Radial blanket and H/D=1.5	Radial blanket and H/D=1.5
Power [MW <sub>th</sub> ]	100	200	100	200	100	200
Initial <sup>233</sup> U load [kg]	654	654	667	667	677	677
Fuel reprocessing of 1I/day						
Feeding in <sup>233</sup> U [kg/an]	11.38	23.38	1.72	4.70	-0.07	0.98
Breeding ratio	-29.83%	-30.64%	-4.52%	-6.16%	0.18%	-1.29%
Total <sup>233</sup> U needed [kg]	1013.87	1388.37	738.83	835.16	715.05	754.25
Breeding ratio (radial + axial fertile blankets)			1.81%	-0.04%		
Fuel reprocessing of 4I/day						
Feeding in <sup>233</sup> U [kg/an]	11.20	22.58	1. 48	3.58	-0.38	-0.26
Breeding ratio	-29.37%	-29.59%	-3.88%	-4.69%	1.00%	0.34%
Total <sup>233</sup> U needed [kg]	1001.86	1353.13	722.50	794.21	709.74	723.03
Breeding ratio (radial + axial fertile blankets)			2.49%	1.54%		

Addition of a radial fertile blanket + Elongated core ⇒ small modular breeder MSFR

### COUPLE code

# Thermo-hydraulic model

The control equations for the liquid-fuel in the COUPLE code are written as following:

Mass conversation equation:

<del>P</del>+V(**b**)=(

Momentum conversation equation:



Energy conversation equation:



#### See the previous presentation:

ZHANG D., ZHAI Z.-G., CHEN X.-N., WANG S., RINEISKI A., "COUPLE, a coupled neutronics and thermal-hydraulics code for transient analyses of molten salt reactors"

# COUPLE code

# Neutronics model

- based on the multi-group (here 2) diffusion theory while considering flow effects of the liquid-fuel

Diffusion equation for the neutron flux of group g:



The balance equation for the delayed neutron precursor of family i:

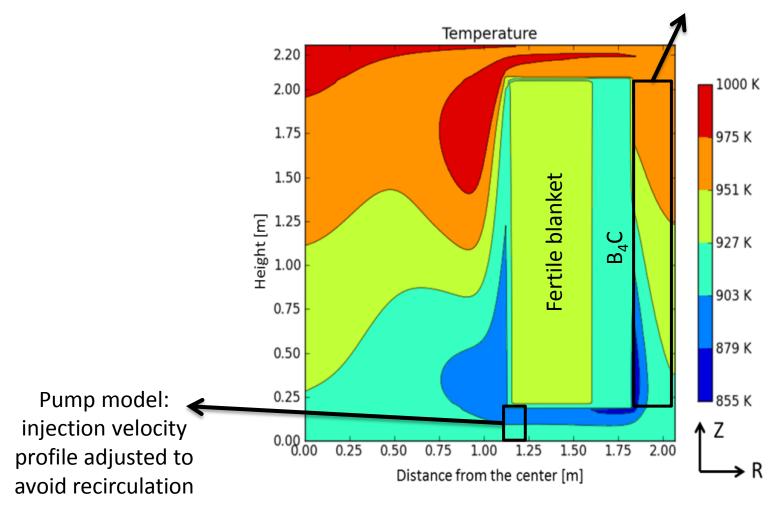


# MSFR model

# Steady state calculation

- Half of the core model
- with 112/130 cells in the R/Z directions

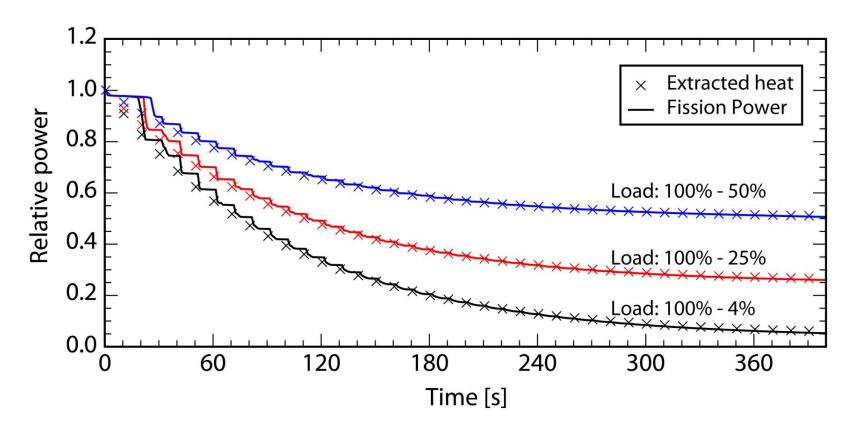
Heat exchanger model: Negative heat source

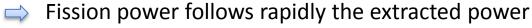


# Operational transients

#### Load following

- negative heat source in the heat exchanger decreases from 100% to 50/25/4% exponentially (stepwise) with  $\tau$ =100s



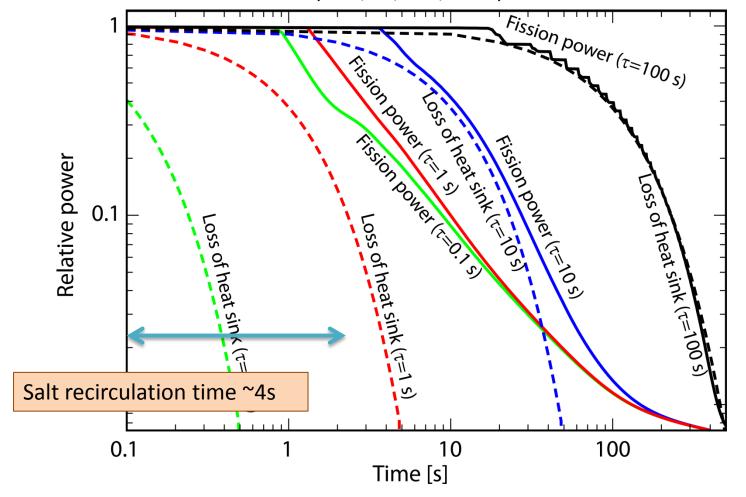


#### Loss of Heat Sink

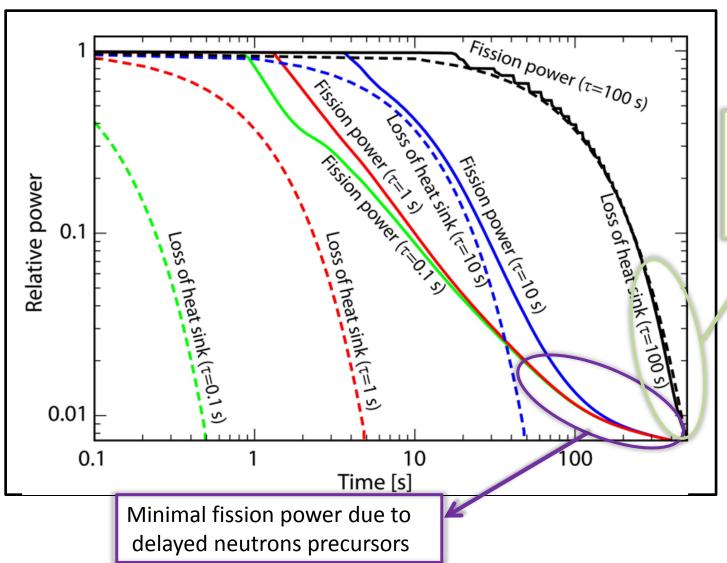
- Fuel salt circulation fixed
- Extracted heat decreases from 100% to 0
- Exponential decrease
- Different inertia are studied (0.1s, 1s, 10s, 100s)



Extracted Power



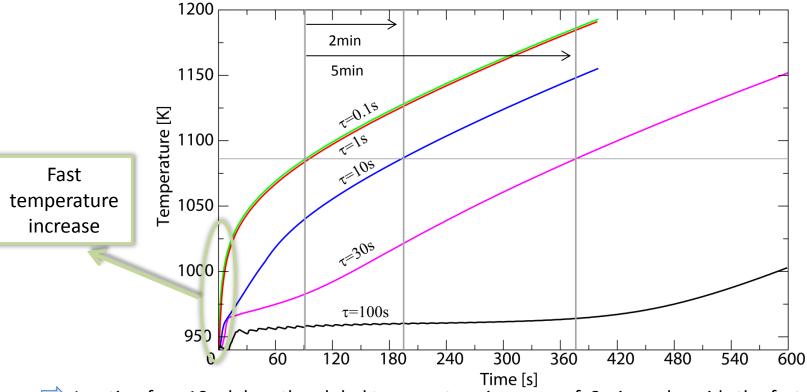
#### Loss of Heat Sink



 $P_{fission} < P_{ext}$ because  $P_{core} = P_{fission} +$ Decay Heat

#### Loss of Heat Sink

- Temperature increase caused by not extracted fission power + decay heat
- → Very pessimistic hypothesis of extracted heat = 0 (heat losses through structure material, natural circulation of the fuel salt and intermediate fluid ...)



- Inertia of  $\tau$  = 10s delays the global temperature increase of 2min and avoids the fast temperature increase
- Inertia of  $\tau$  > 10s should be implemented on the pumps of the fuel circuit and the intermediate circuit

How to manage this temperature increase?

- Protection systems in the fuel salt circuit studied (redondant safety cooling system or natural convection)

