Molten Salt Reactor Technology

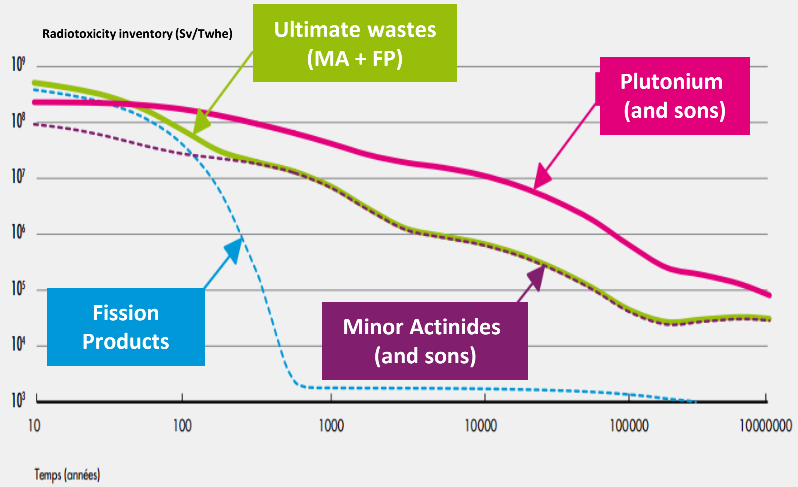
Opportunities of molten salt fuel for actinides management

CEA
P. ALLEGRE, C. CHABERT, P. GAUTHE, E. MARTIN-LOPEZ, J. MARTINET, L. MATTEO, M. MASCARON, V. PASCAL, J. SERP, L. TARDIEU, C. VENARD

ORANO
B. MOREL, I. MORLAES, G. SENENTZ, F. SOUVIGNET

- **French reactor fleet : Light water reactors with a mix of enriched uranium (UOX) and plutonium (MOX) fuels**
- **Spent fuel reprocessing strategy : "Pu valorization"**
  - Pu from UOX reprocessing is used in LWR (MOX fuel strategy)
  - Pu is the main transuranic actinides produced and the main contributor to the long term radiotoxicity
  - ⇒ **Mono-recycling strategy (Uranium savings, Waste management)**
- **In LWR (thermal spectrum effect) : Pu quality decreases and minor actinides production is enhanced**
  - Fissile quality of Pu extracted from used MOX fuel is degraded and MA content is upper than in used UOX fuel
  - Americium is a major contributor to the long term radiotoxicity and thermal load of wastes (high impact on waste storage facility volume)
  - ⇒ **Multi-recycling strategy of Pu is an open issue for LWR (R&D) + LWR not adapted to convert MA**

Radiotoxicity evolution of an UOX fuel (45GWd/t)



Reference : Séparation-transmutation des éléments radioactifs à vie longue, CEA, 2012

Minor actinides inventories in used fuel (after a 5 year cooling time)

Isotopes	UOX -REP 46 GWj/t (g/TWhe)	MOX-REP 48 GWj/t (g/TWhe)
^{237}Np	1 700	390
Total Np	**1 700**	**390**
^{241}Am	1 160	8 900
^{243}Am	540	5 100
Total Am	**1 700**	**14 000**
^{244}Cm	190	2 400
^{245}Cm	16	420
Total Cm	**210**	**2 900**
Total AM	**3 600**	**17 000**

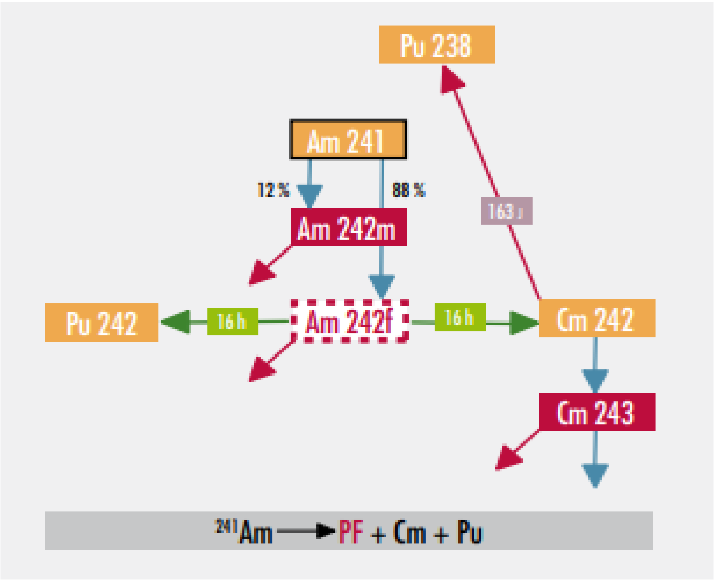
PLUTONIUM MANAGEMENT & TRANSMUTATION IN FAST REACTORS

- **Fast spectrum systems are well adapted to transuranic actinides transmutation by fission process**
  - High energy neutrons **enhance Pu regeneration cycle**
  - High energy neutrons **enhance transuranic fission reactions** (cf. threshold fissions)
  - High energy neutrons **minimize transuranic capture reactions** responsible of minor actinides
  - ⇒ **Fast spectrum : Stabilization of Pu inventory without quality degradation, weaker production of MA**
  - ⇒ Various French programs around SFR technology, Pu management and transmutation since 80s
- **MA transmutation process is a time-consuming phenomena which rely on multi-recycling strategies (10 to 50y)**
  - Only a small part of MA fission at each cycle (weak fission cross sections in fast spectrum)
  - Rely also on capture process for poorly fissile actinides which will form highly fissile actinides (ex: Am^{241} -> Am^{242})
- **One complete cycle time relies on irradiation, cooling, reprocessing and fuel fabrication time => ~ 15 years / cycle**

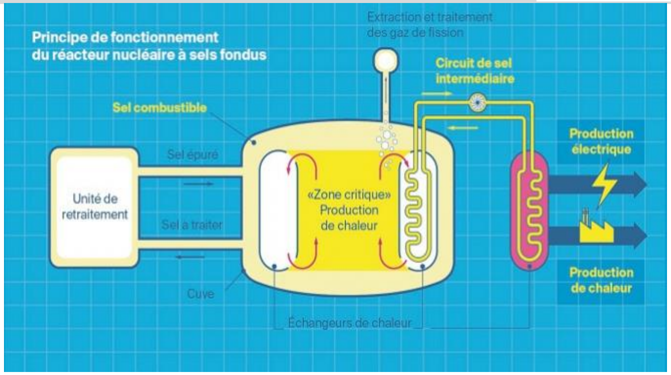
MINOR ACTINIDES INVENTORIES IN USED FUEL (AFTER A 5 YEAR COOLING TIME)

Isotopes	MOX-REP 48 GWj/t (g/TWhe)	MOX-RNR 99 GWj/t (g/TWhe à l'équilibre)
^{237}Np	390	460
Total Np	**390**	**460**
^{241}Am	8900	2900
^{243}Am	5100	680
Total Am	**14000**	**3600**
^{244}Cm	2400	190
^{245}Cm	420	18
Total Cm	**2900**	**215**
Total AM	**17000**	**4300**

Reference : Séparation-transmutation des éléments radioactifs à vie longue, CEA, 2012

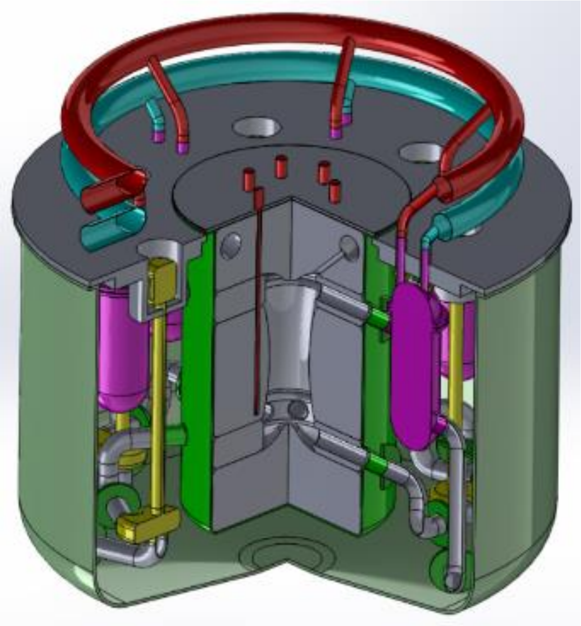


- **Molten salt reactor technology**
  - A high temperature liquid fuel flowing in a loop with no pressure
  - High thermal safety feedback effects (fast liquid thermal expansion)
  - Potential high source temperature for energy conversion systems
  - **Needs to manage fission gas and salt compositions**
  - **Technological issues requiring R&D steps (materials, corrosion …)**



@Crédit : CNRS

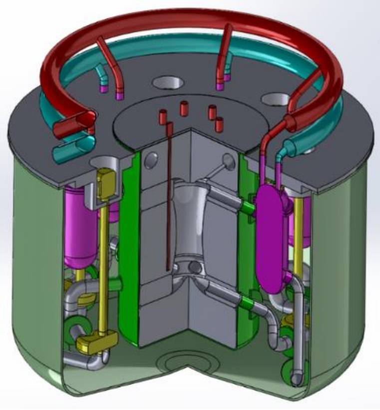
- **Can Fast MSR enhance plutonium management or transmutation ?**
  - Irradiation times no more limited by clad ageing
    => opportunity to increase fuel in-core irradiation time
  - Recycling does not need solid to liquid process and could use high temperature and radioactive recycling process (pyrochemistry)
    => opportunity to reduce cooling and reprocessing time
  - Unlike in SFR, MA do not degrade safety feedback effects
    => opportunity to increase molar fraction of actinides
  - Fast MSR easily accept various kind of isotopic vector
    => opportunity to increase fuel management flexibility
  - Last but not least, **opportunity to harden neutron spectrum** by removing oxygen present in current ceramic fuel



@Crédit : CEA, ARAMIS

- **Which goals for a Fast Molten Salt Reactor with respect to bad quality plutonium coming from LWR MOX used fuel ?**
  - **Full converter mode (Plutonium, Americium …)**
  - Breeder/Isogeneration mode for Pu and Burning mode for Am
  - Help to launch new regeneration cycle (Thorium cycle)

- **French context with respect to the « Full converter mode »**
  - **2020 – 2022 : The ARAMIS project (CEA/ORANO)**
    - Small plutonium molten salt convertor
    - Preliminary design studies and 1st chemical process studies
    - Power of 300 MWth – Consumption of ~ 130 kg Pu/y
  - **2022 – 2025 : The ISAC project (CEA/CNRS/EDF/FRA/ORANO)**
    - Americium and Plutonium molten salt transmuter
    - Scenarii studies, preliminary design, 1st experiments (material corrosion, salt properties measurements, salt loops)
    - Context : French 2008 Law concerning the sustainable management of nuclear materials and wastes



@Crédit : CEA, ARAMIS

@Crédit : CEA, PuCl3 synthesis

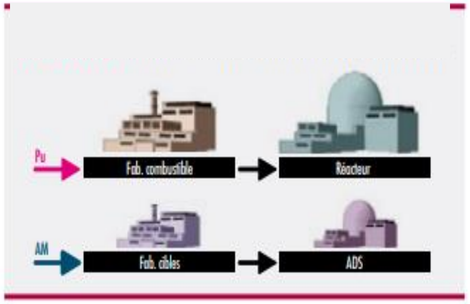
- **Public Technical Report on the sustainable management of nuclear materials**
  - **Issued by CEA in december 2012 with respect with the 2008 French Law**
  - Various contributors : Andra, CEA, CNRS, French universities, AREVA, EDF
  - Goal : Evaluation of industrial perspectives of separation and transmutation of long lived radioactive isotopes as an alternative to waste storage

- **Evaluation of various Gen IV technologies to incinerate actinides (from Pu to Cm)**
  - Main technologies assessed : **SFR and ADS** (+ *quick evaluation of GFR, LFR and MSR*)

Comparison of MA transmutation efficiencies of ADS and SFR technologies

MODE DE TRANSMUTATION	Homogène			Hétérogène (CCAM)		ADS
Teneur AM	1 %	2 %	4 %	10 %	20 %	55 %
Capacité de transmutation AM (kg/TWhe)	0*	5*	14*	3,5 (-0,5*)	6 à 8 (2 à 4*)	95*

* capacité de transmutation d'actinides mineurs exogènes au réacteur



Reference : Séparation-transmutation des éléments radioactifs à vie longue, CEA, 2012

  - **Best MA transmutation efficiency in solid fuel concept : the ADS (thanks to high MA content)**
  - **Opportunity for Fast MSR to combine high MA content and liquid fuel advantages (no limitations due to clad irradiation, no solid fuel fabrication, less time consuming recycling processes)**

- **MA production of French reactor fleet (380 Twhe) :**
  - Data coming from "Dossier 2012"

Production of Minor Actinides	
LWR fueled with Uox	~ 1,3 t/y
LWR fueled with MOX	~ 6,5 t/y
SFR fueled with (ex)MOX – equilibrium state	~ 1,4 t/y

Reference : Séparation-transmutation des éléments radioactifs à vie longue, CEA, 2012

- **Evaluation of number of dedicated reactors :**
  - Hypothesis of "Dossier 2012" : 2 t/y
  - ADS 385 MWth "Dossier 2012" : 16 units
  - **MSR with P > 1500 MWth and transmutation efficiency > 50 kg/Twhe : < 10 units**
  - **/!\ Quantities of Pu and Am in the reactor should be reasonable !**

Number of reactor to transmute 2t/y of MA			
	50 kg/TWhe	75 Kg/TWhe	95 kg/Twhe
300 MWth	39	26	21
1500MWth	8	5	4
3000MWth	4	3	2

Plutonium hold up in reactor (t)	
ADS 385 MWth *(« Dossier 2012 »)*	~ 2,5 t
ARAMIS 300 MWth *(« Preliminary evaluation 2021»)*	~ 3,5 t *(0,9 t in core active zone)*
MSR 1500 à 3000 MWth	??

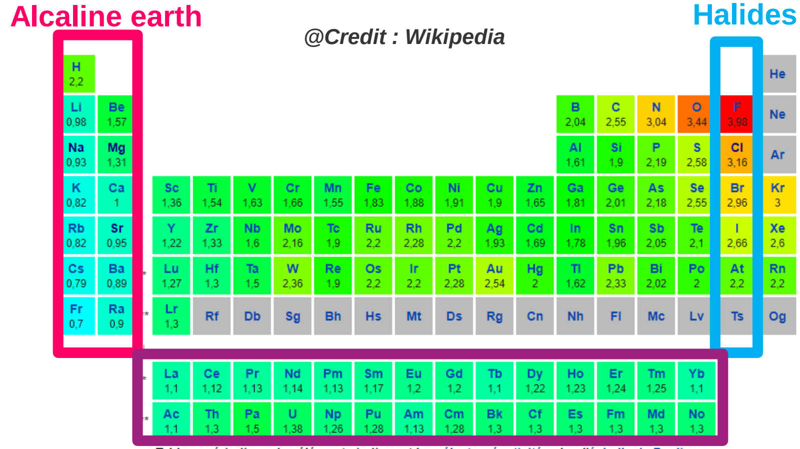
- **Goal of ISAC project : Perform an evaluation of MSR potential with respect to transmutation of MA and compare it to "Dossier 2012" scenarii studies**

MOLTEN FUEL SALT CHOICE FOR A TRANSMUTER CONCEPT

- **Base elements greatly depends on reactor objectives (transmuter, breeder …) and nuclear cycle (thorium/plutonium)**

- **Various family of halides salts according the anions : F, Cl, Br, I …**
  - I and Br disqualified (low knowledge, too high capture XS)

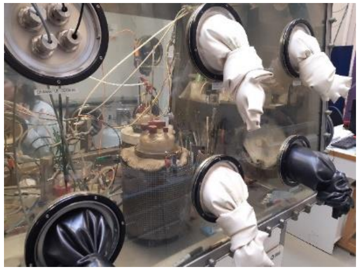
- **Criteria to choose base salt :**
  - High solubility of fissile and target elements (Pu, Am)
  - Compatibility with available recycling process (hydro/pyro)
  - Low melting temperature
  - Low moderation induces by base elements
  - Salt stability under irradiation (low radiolysis, low activation, low production of corrosive elements for structures)
  - Good thermal properties (thermal capacity, density …)
  - Good behavior in case of external aggression (O_2/water leaks, secondary coolant leak …)



@Credit : Wikipedia

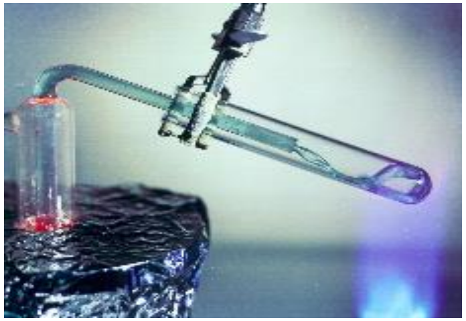
Tableau périodique des éléments indiquant leur électronégativité selon l'échelle de Pauling

Actinides et Lanthanides



@Credit : CEA Marcoule, chloride salts

@Crédit : Los Alamos, chloride salt



@Crédit : Wikipedia, Molten FLiBe

MOLTEN FUEL SALT CHOICE FOR A TRANSMUTER CONCEPT

	Fluorides		Chlorides	
Neutron spectrum	Softer	✗	Faster	✓
Solubility of Pu	Low solubility	✗	Higher solubility	✓
Melting temperature	(often) Higher	✗	(often) Lower	✓
Boiling temperature	(often) Higher	✓	(often) Lower	✗
Coolant properties	(often) Better Better experimental knowledge	✓	(often) Lower Lack of experimental data	✗
Chemical process R&D feedback	R&D scale only	✗	Reprocessing metallic fuel (INL) Reprocessing oxyde fuel (RIAR)	✓
Hydro reprocessing compatibility	No	✗	Yes	✓
Operational Feedback	ARE, MSRE, Test loop facilities	✓	No operational feedback Test loop facilities (TerraPower) On going project : MCRE 2025 (Terrapower)	✗

- Chlorides salts appears to be well adapted to :
  - *Fast spectrum concept*
  - High actinides content (Pu, Am)
  - Compatible with hydro reprocessing

But requires more technological and base R&D programs to increase reactor TRL

No operational feedback !

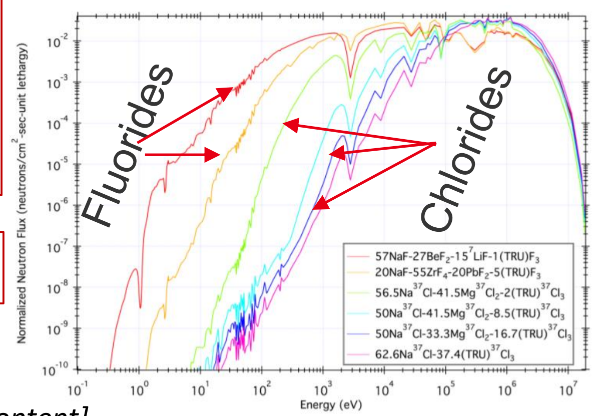
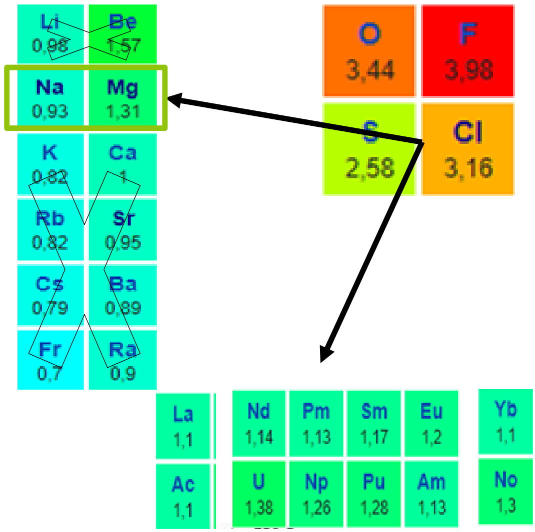


Fig. 3. FS-MSR TRU burner spectra.

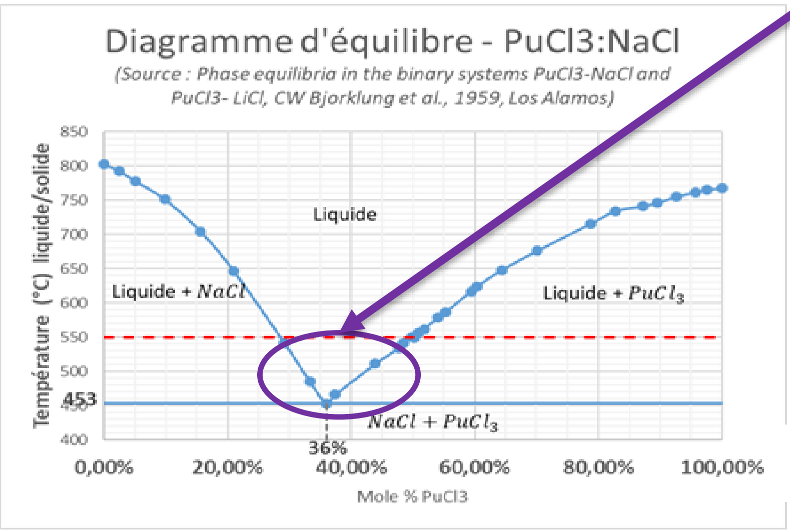
- **Cl salts for fast MSR project : Terrapower (USA), Elysium (USA), MOLTEX (GB)**
- **F salts for fast MSR project : MOSART (Russia)** *[= transmuter with very low actinide content]*

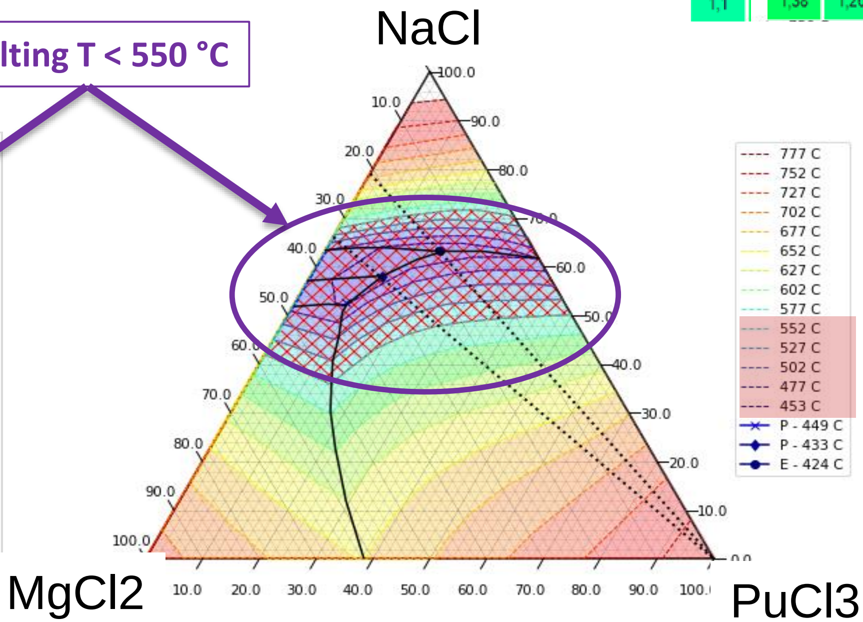
- **Salt elements behavior under irradiation**
  - Disqualified : Li, Be (moderator), K and Ca (production of $^{40}K/^{36}Cl$ and $^{40}K/^{41}Ca$)
  - **Na and Mg = best candidates (no moderation, low capture, no long lived el.)**
  - Chlorine has to be enriched in ^{37}Cl (between 70% to 99%) to limit ^{36}Cl and S



- **Salt melting temperature : Binary/ternary mix reduces melting temperature**
  - **For a given upper limit in melting temperature, ternary mix are far more flexible in terms of actinide contents**
    => bigger liquid domain

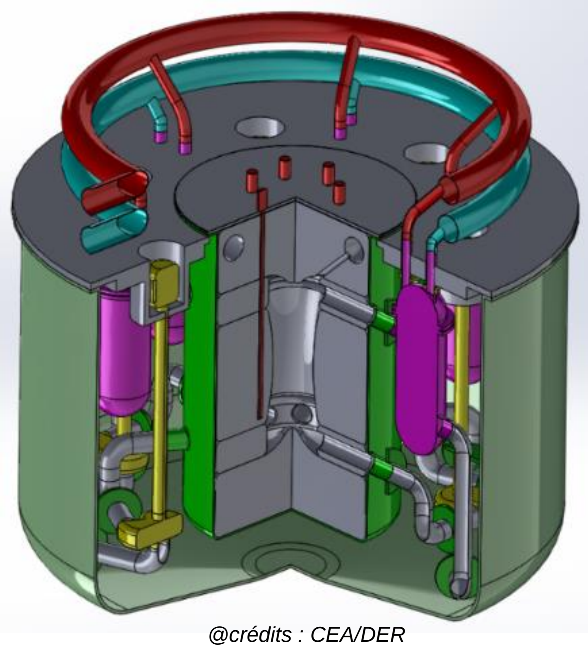
Melting T < 550 °C





@Source: Thermodynamic evaluation of the NaCl-UCl$_3$-PuCl$_3$-MgCl$_2$ system, O. Benes, RJM. Könings, Journal of nuclear material, 2008

- **The ARAMIS Project : A small fast neutron actinide convertor (Pu, Am)**
  - Ternary chloride salt without uranium ($NaCl-MgCl_2-(Pu,Am)Cl_3$)
  - Enriched chloride (^{37}Cl) to prevent excessive formation of ^{36}Cl and S
  - Loop type reactor confined in a vault security vessel
  - Magnesium oxyde reflectors with boron carbide neutron protection
  - Reactivity control devices
  - High performance shell and Tubes heat exchangers
  - Small core active zone with respect to total fuel volume (25%)



@crédits : CEA/DER

- **ARAMIS transmutation performance**
  - Preliminary results to consider with caution (start of ISAC project)
  - At least 5 mol% of AmCl3 seems needed to reach 50 kg/Twhe
  - **Production of Cm**

P = 300 MWth	ARAMIS Pu	ARAMIS Pu+ 2mol% Am	ARAMIS Pu+ 5mol% Am	ARAMIS Pu+ 8mol% Am
$PuCl_3$ (mol%)	15	15	13	12
$AmCl_3$ (mol%)	/	2	5	8
Pu (kg/Twhe)	-114	-103	-76	-64
Am (kg/Twhe)	+6	-11	-52	-73
Cm (kg/Twhe)	+0,5	+7	+26	+34

[1] based on preliminary evaluation - @crédits : CEA/DER/SPRC

- **French nuclear fuel and waste management strategy**
  - Treatment of UOx spent fuel and Pu recycling in LWR already implemented (MOX strategy)
    « Plutonium valorisation » = Uranium savings, Waste Storage Surface limitation
    Plutonium quality drop and enhanced production of americium when multiple recycling in LWR
  - Treatment of MOX spent fuel and Pu multiple recycling is studied as a medium (LWR) and long term objective (Fast reactors)
- **Fast Molten Chloride Salt Reactor has the potential to use degraded Pu from MOX (as SFR)**
  - Opportunities to enhance transmutation (in quantity and in processing time)
  - A lot of scientific and technical challenges : chloride salts are badly knowns, salt depletion, reprocessing strategy, fission products behaviour, material corrosion, thermal and irradiation damages, components handling, monitoring …
  - New safety guidelines to invent
- **ISAC Project aims to assess the potential of fast MSR to enhance French nuclear material management strategy**
  - « France 2030 » program with 5 partners (CEA/CNRS/EDF/FRAMATOME/ORANO)
  - Scenarii studies to assess the potential benefits of MSR with respect to the French nuclear cycle
  - Preliminary reactor design and operating studies to consolidate the concept
  - First batch of experimental studies (salt synthesis, salt property measurements, salt loop, corrosion studies)

THANK YOU FOR YOUR ATTENTION