Research of Thorium Uranium Fuel Cycle in TMSR

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TMSR 钍基熔盐核能系统

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Shanghai Institute of Applied Physics, Chinese Academy of Sciences

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OUTLINE

Motivation of TMSR Th-U fuel cycle

Advanced Th-U Fuel Cycle strategy

Research progress of TMSR Th-U fuel cycle

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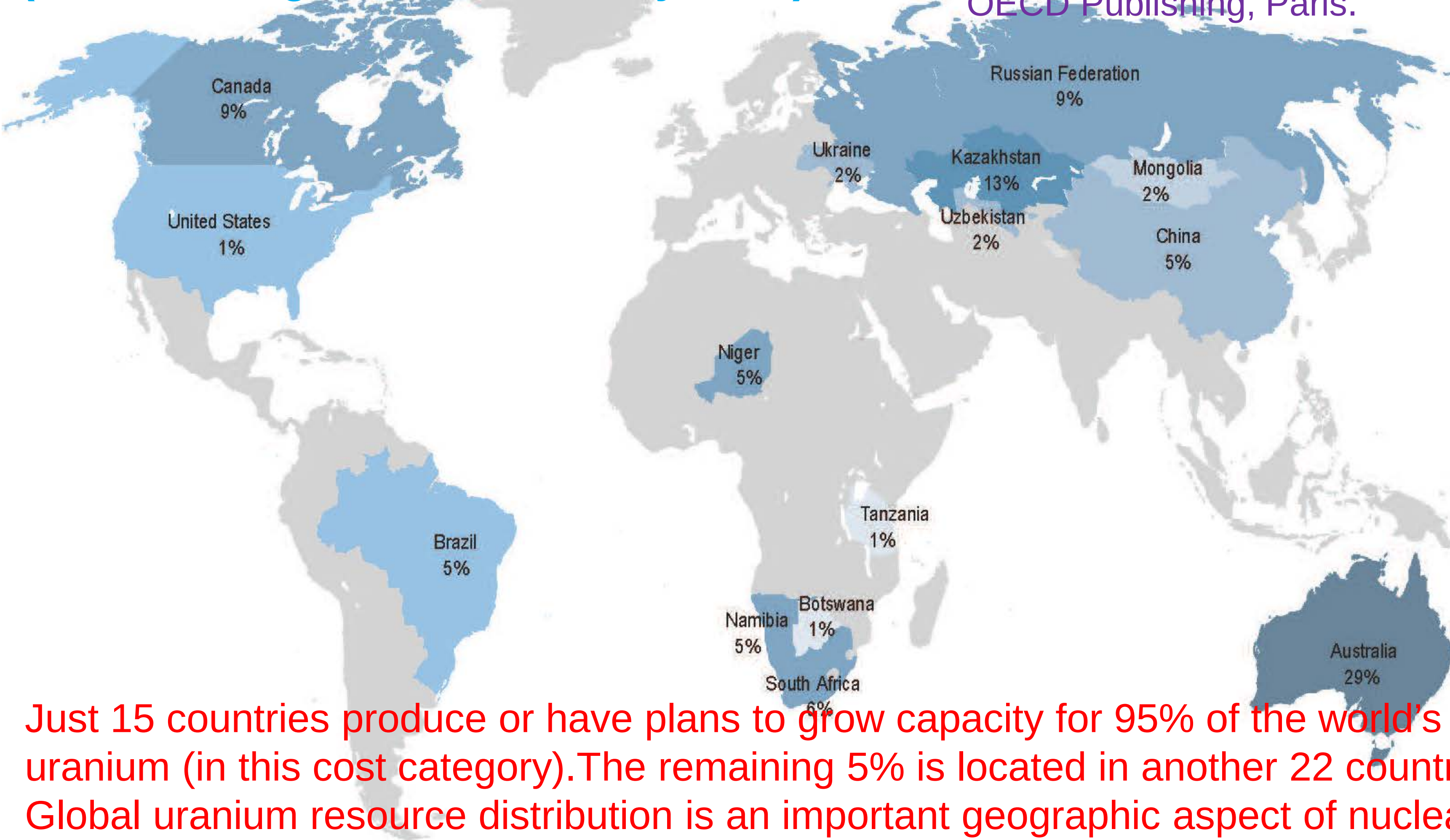
Global Uranium Reserves

Global distribution of identified resources (<USD 130/kgU as of 1 January 2015)

Source：NEA (2016), Uranium 2016: Resources, Production and Demand, OECD Publishing, Paris.



Just 15 countries produce or have plans to grow capacity for 95% of the world's uranium (in this cost category).The remaining 5% is located in another 22 countries. Global uranium resource distribution is an important geographic aspect of nuclear energy in light of energy supply security.

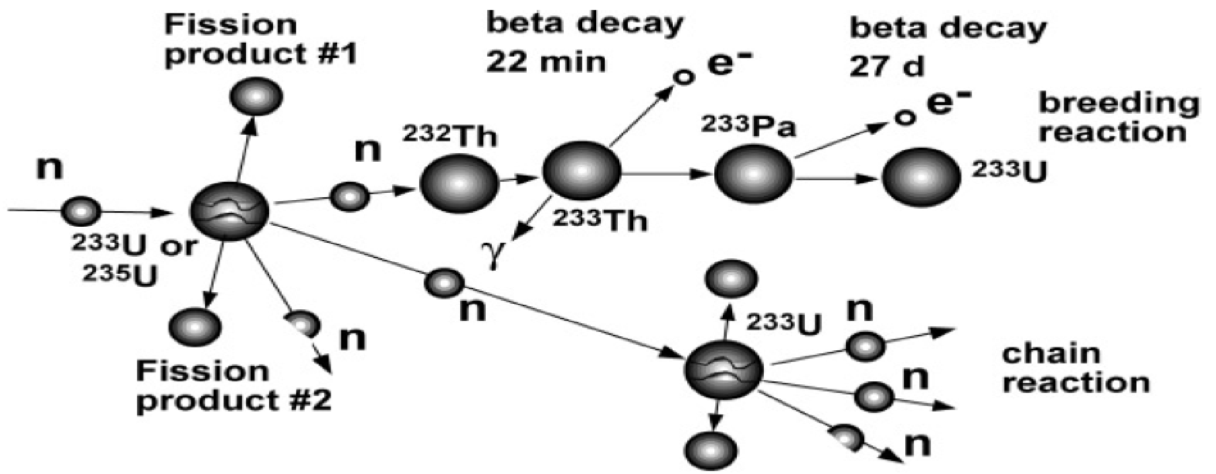
Thorium Reserves in China

- **US Geological Survey: 1-10 million tons**
  - China's National Bureau of Statistics predicted that the rare earth reserve in China is around 18.6 million tons, and US Geological Survey predicted it as 89 million tons. The thorium content in rare earth is around 2-22%
- **Thorium in coal is around 62 million tons**
  - China's coal reserves is around 5.9 trillion tons, with thorium around 62 million tons.
- **Thorium in fly ash is around 6,200 tons per year**
  - Electricity generated from coal in China is around 4.9 trillion kWh in 2018, with 170 million tons fly ash produced and ~6000 tons thorium alongside.

By well optimized Thorium reactors, Thorium from China's REE basic reserves are adequate to produce today China's electricity (3.2 Trillion kWh) for ≈20'000 years ! ***—1984 Nobel Prize winners in physics, Prof. Carlo Rubbia***

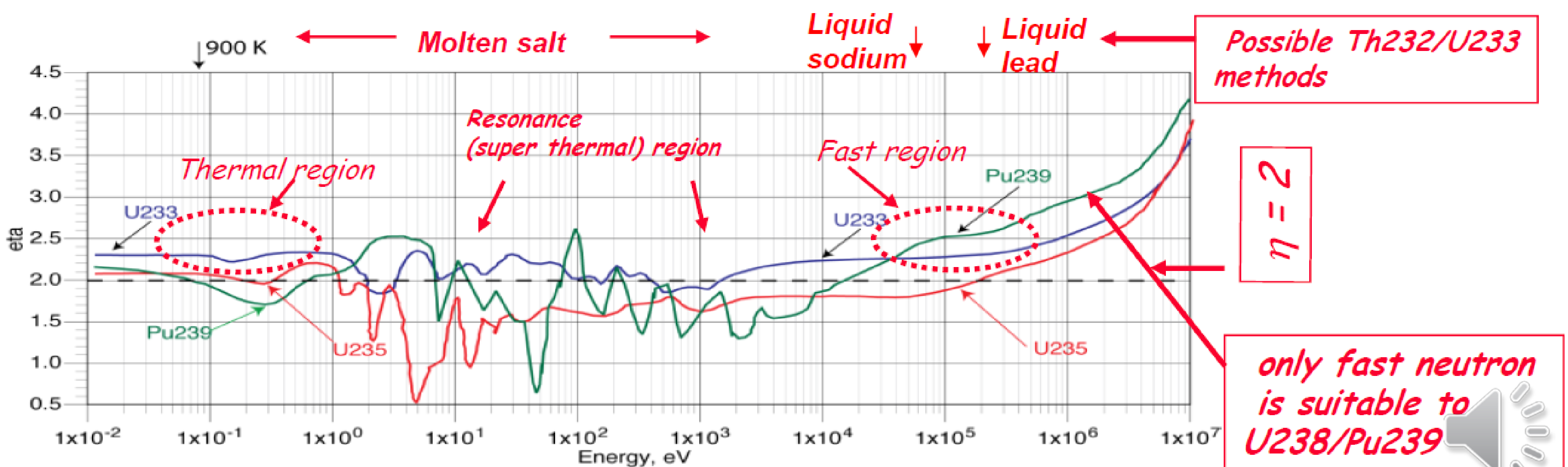
[1] C. Rubbia, "A future for thorium power?" Oct. 27-31, 2013
[2] IAEA, Uranium 2016: Resources, Production and Demand

Th232/U-233 and U238/Pu239 fuel cycles

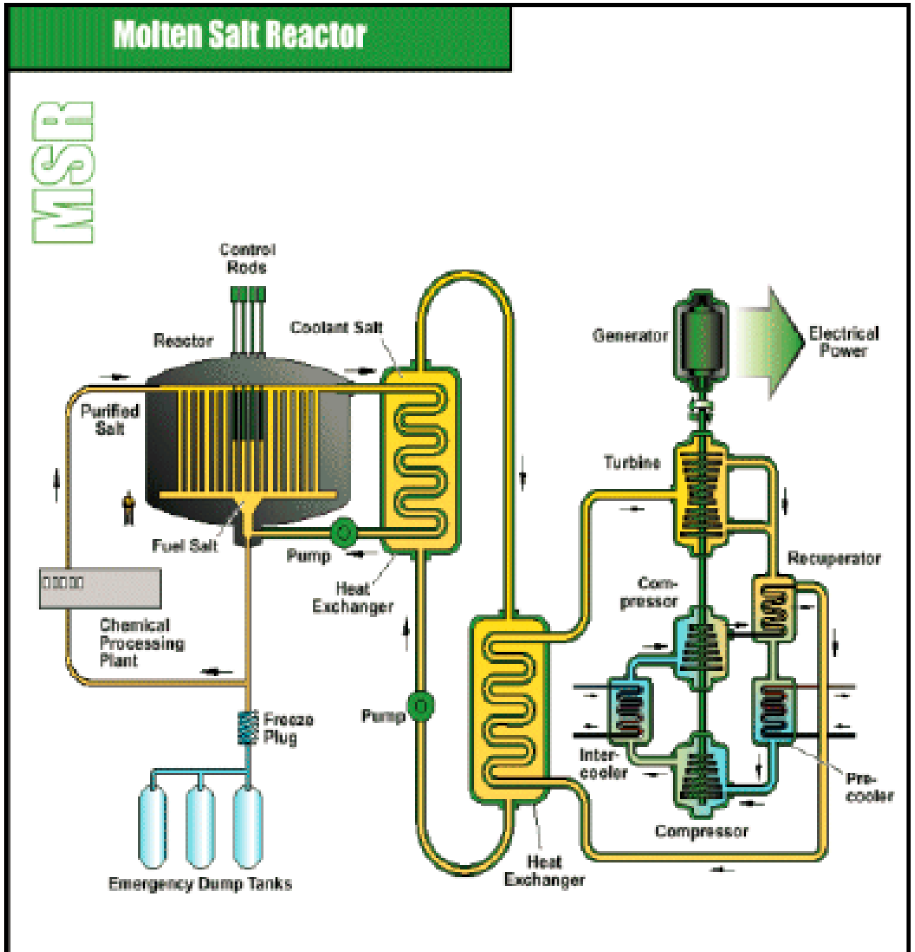


Mean released neutron number per fission η

η = 2 is the required condition for a sustain reactor



Molten Salt Reactor (MSR): desirable for Th utilization



- Specific features of MSR comes out from the use of liquid fuel circulating in the primary circuit
- Typical fuel: actinide fluorides dissolved in fluoride carrier(^7LiF-BeF_2)
- On-line processing of fuel without shutting down of reactor

On-line extracted from core

$$^{232}\mathrm{Th}(\mathrm{n},\gamma) \rightarrow {}^{232}\mathrm{Th}(\beta^-; 22\ \mathrm{min}) \rightarrow {}^{233}\mathrm{Pa}(\beta^-; 27\ \mathrm{days}) \rightarrow {}^{233}\mathrm{U}$$

TMSR Project (Chinese Academy of Sciences)

中文名称：钍基熔盐堆核能系统

英文名称：Thorium Molten Salt Reactor

Nuclear Energy System

Abbr.　：TMSR

Aims　：Develop Th-Energy,　Non-electric application of Nuclear Energy based on TMSR during coming 20-30 years.

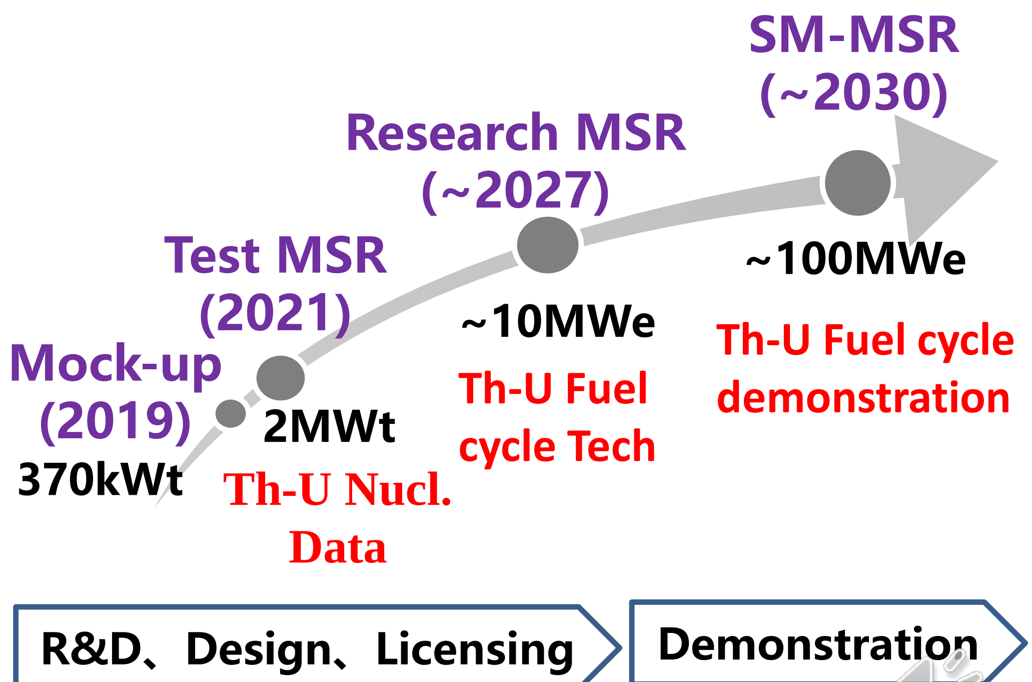
Initiated by Chinese Academy of Sciences (CAS) in 2011

Th-U fuel cycle in TMSR project

Developed key technologies of Th-U fuel cycle, realizing large-scale thorium utilization on small modular MSR (SM-MSR)

Mock-up facility in SINAP 2019

TMSR road map



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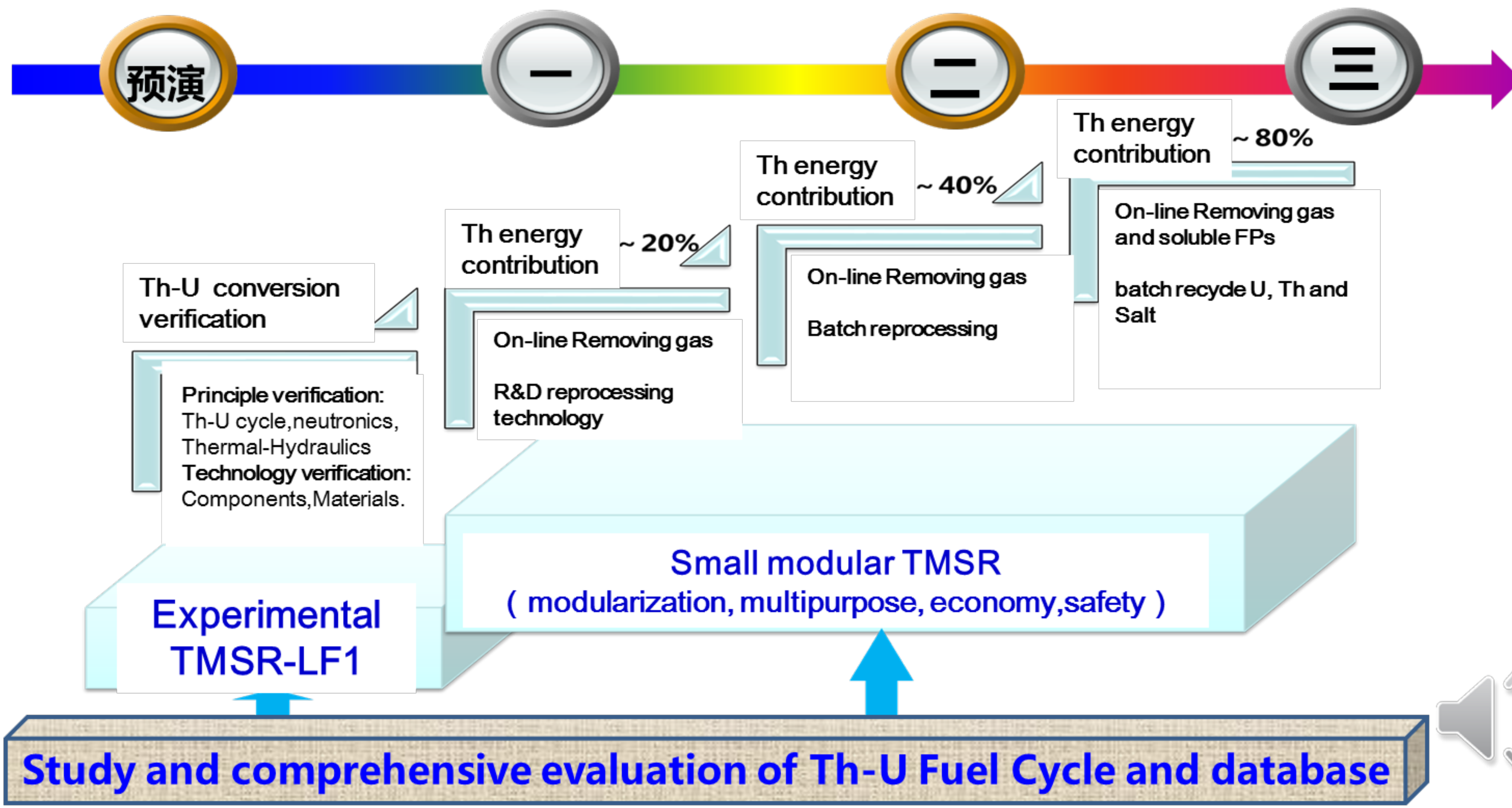
TMSR's Th-U fuel cycle strategy consideration

- **Goal:** Improving thorium resource utilization, minimizing nuclear waste, and promoting nuclear nonproliferation
- **Fuel cycle system:** Small modular MSR, Online removing gaseous FP, Offline fuel reprocessing, Fuel salt reconstruction and/or Online removing non-gaseous FP
- Considering the technology readiness especially the fuel pyro-reprocessing, we propose a "Three-step" development strategy for thorium utilization in MSR

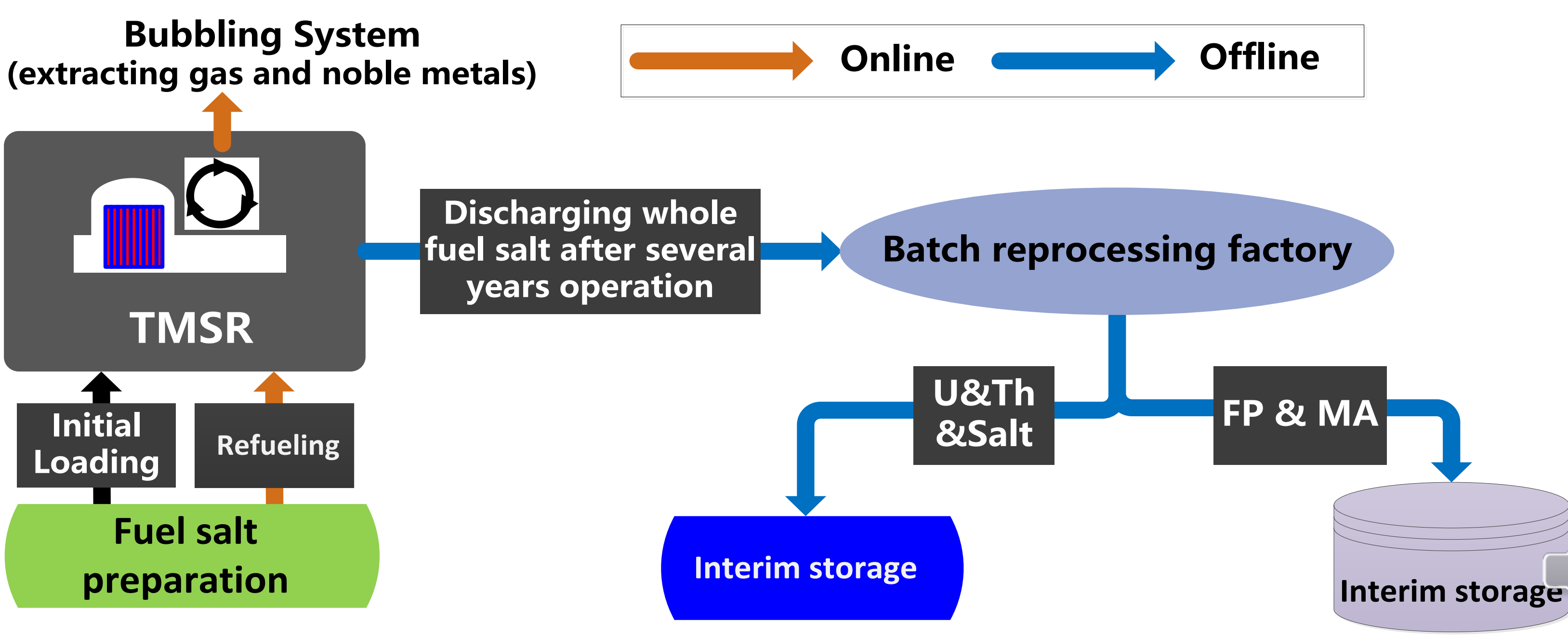
Planed Thorium utilization strategy in TMSR and its performances

Combine the MSR with pyro-processing technology to realize the efficient thorium utilization of small modular TMSR.



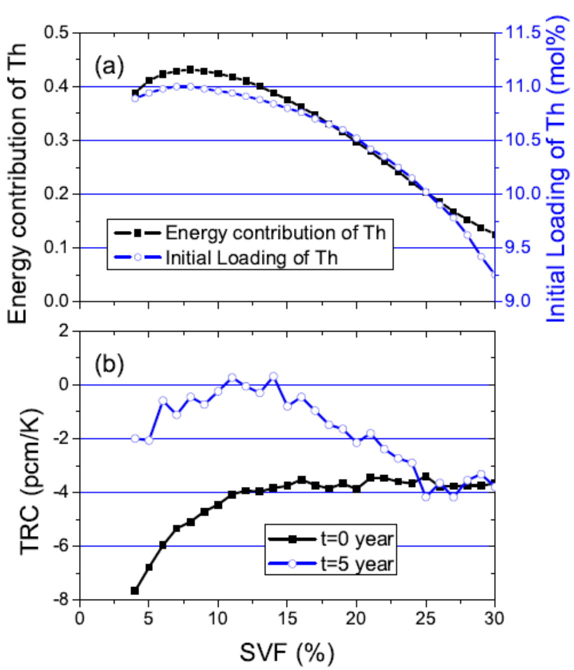
Step 1: Once through mode→Starting Thorium utilization

- **Initial loading :** LEU+Th (Fresh fuel salt)
- **Online refueling**: Fresh fuel salt
- Continuously removing gaseous FPs
- Discharge all fuel salt after several years' operation and extract U and Th for reserve
- And then FP and MA for temporary storage

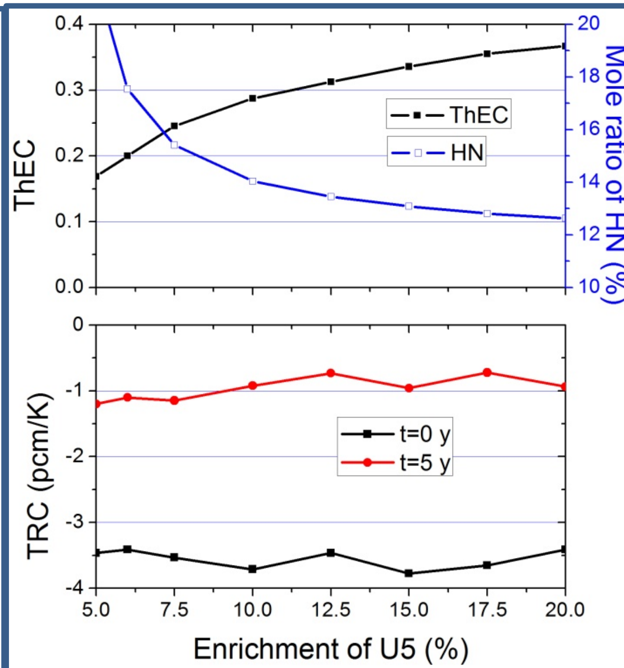


Baseline Salt volume fraction & LEU enrichment optimization

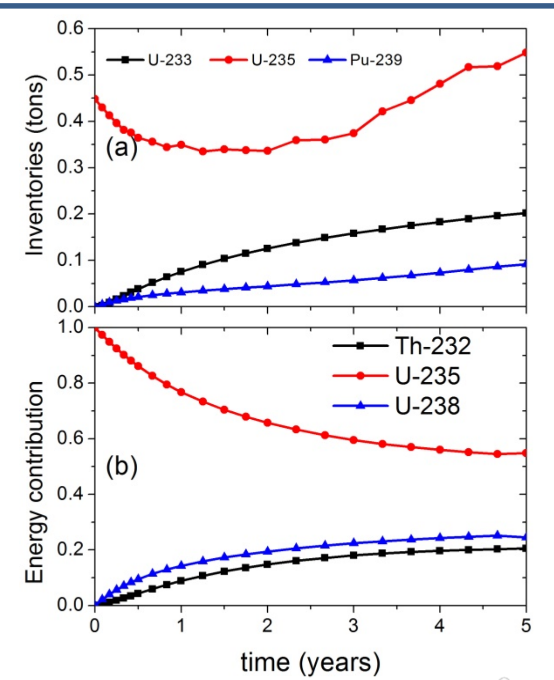
- Enhanced energy contribution from Th due to increased amount of U-233.
- SVF set to 15% considering the demand of large negative TRC.
- U-235 with 6% enrichment to realize the 20% energy contribution considering the higher fuel utilization.



ThEC VS salt volume fraction



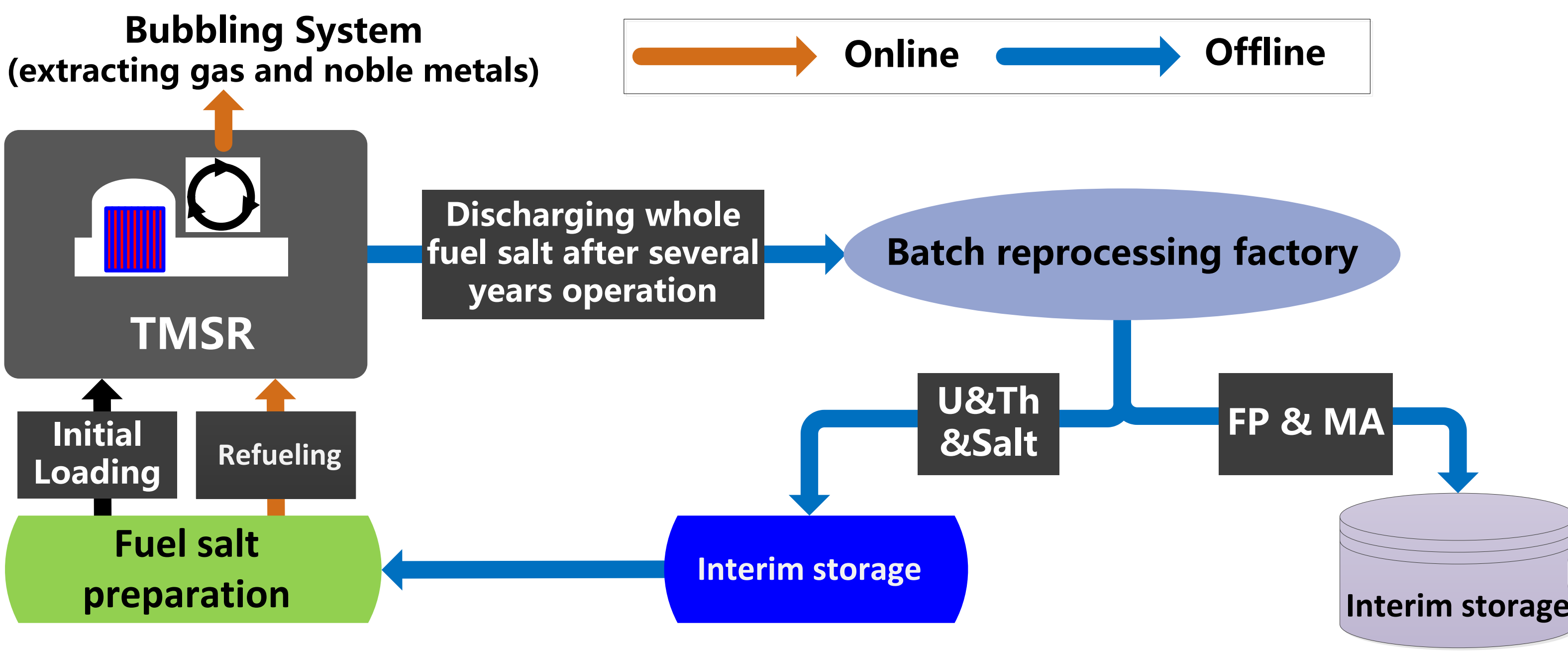
ThEC VS LEU enrichment



Mass and energy contribution

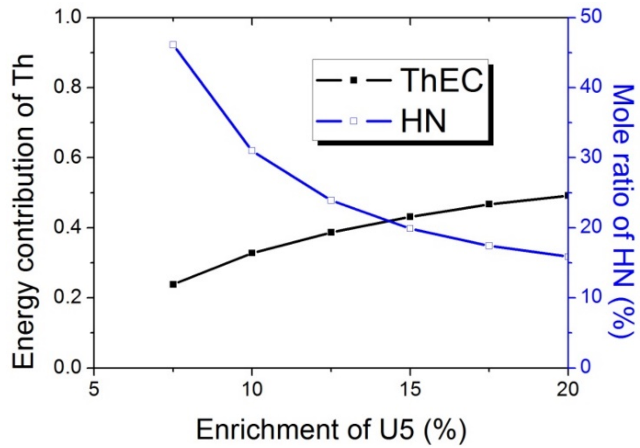
Step 2: Batch reprocessing+Fuel reloading→Th/U transition

- **Initial loading :** LEU+Th (Fresh fuel salt)
- Online refueling: Fresh fuel salt+ Reconstructed fuel salt
- Continuously removing gaseous FPs
- Discharge all fuel salt after several years operation and extract U and Th for reloading
- And then FP and MA for temporary storage

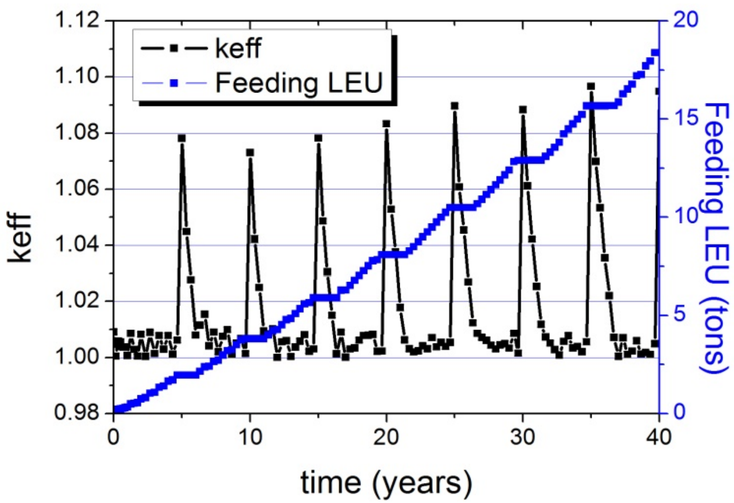


More Energy from Thorium at step 2 by recycling fuel with batch processing

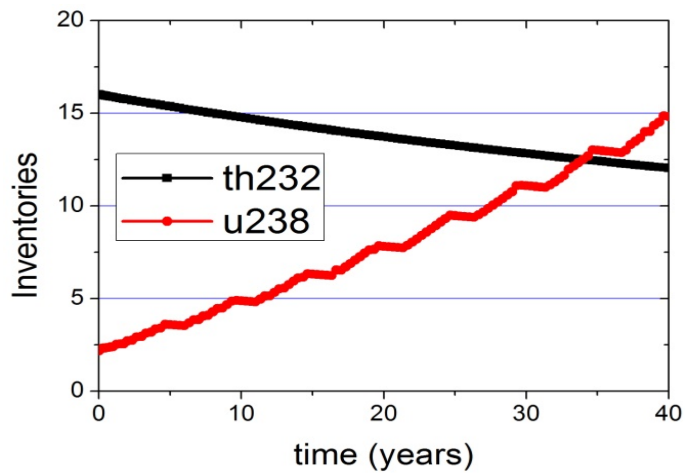
- U5 enrichment should be no less than 17.5% to ensure molten salt chemical stability
- Thorium contribution can achieve about 40%.



ThEC VS U enrichment



Keff and Feeding LEU

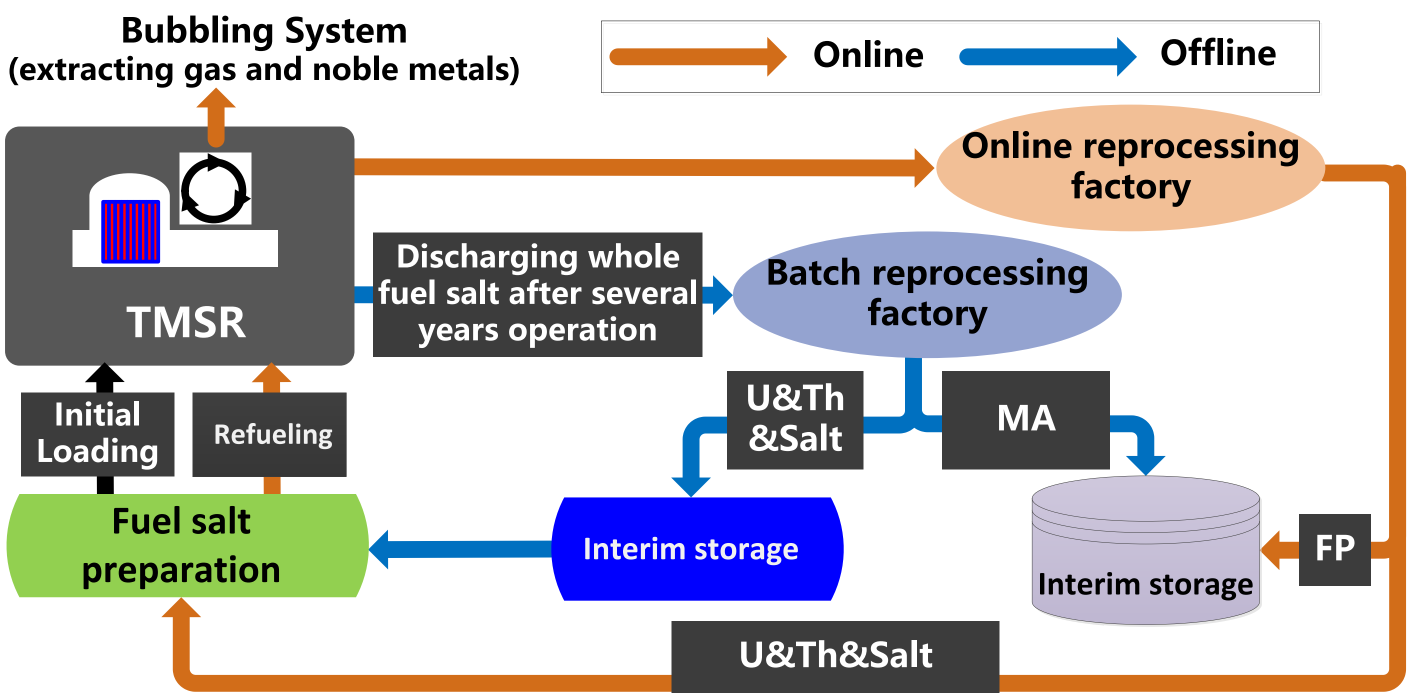


Mass of Fertile

Step 3: Batch reprocessing+Fuel reloading +Online removing FP → Improved Th utilization

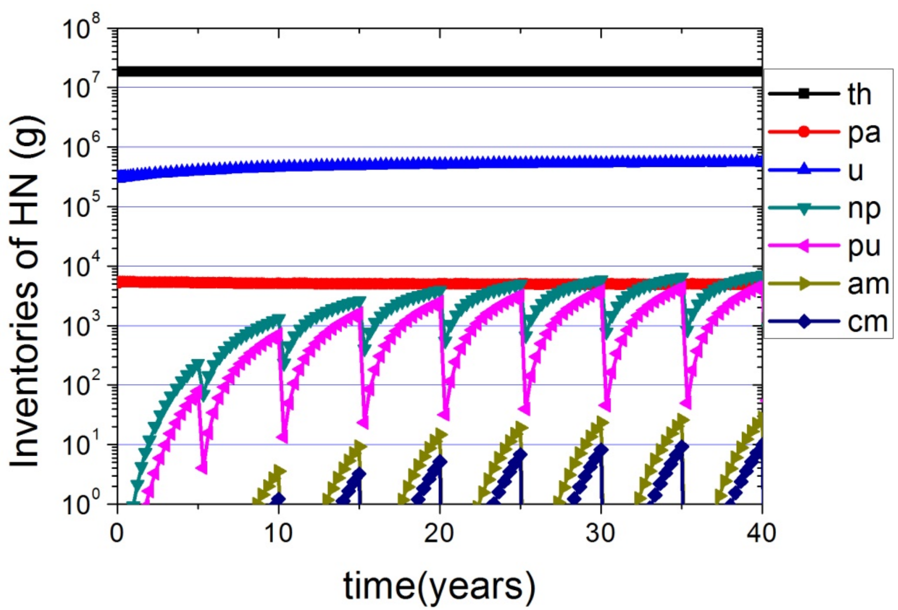
- **Initial loading :** LEU+Th (Fresh fuel salt)
- **Online refueling:** Fresh fuel salt+ reconstructed fuel salt
- Continuously removing gaseous FPs
- Online removing non-gaseous FP

- Discharge all fuel salt after several years operation and extract U and Th for reloading
- And then FP and MA for temporary storage

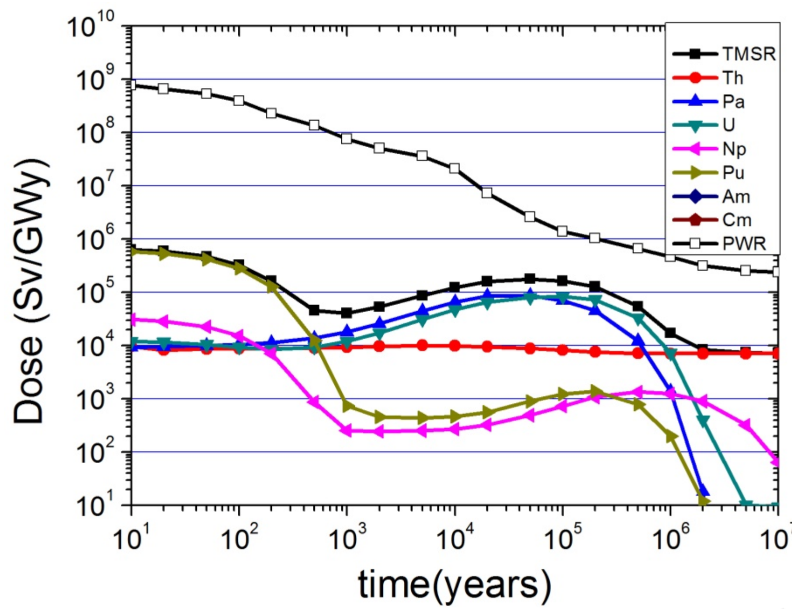


Realizing Large-scale Thorium Utilization

- Achieving about 80% energy contribution from Th during the whole operation.
- Obtaining a much lower radioactivity than that of LWR with enriched uranium.



Evolution of HN



Radioactivity of discharged HN

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Advanced Th-U Fuel Cycle strategy

Research progress of TMSR Th-U fuel cycle

R&D of TMSR Th-U fuel cycle

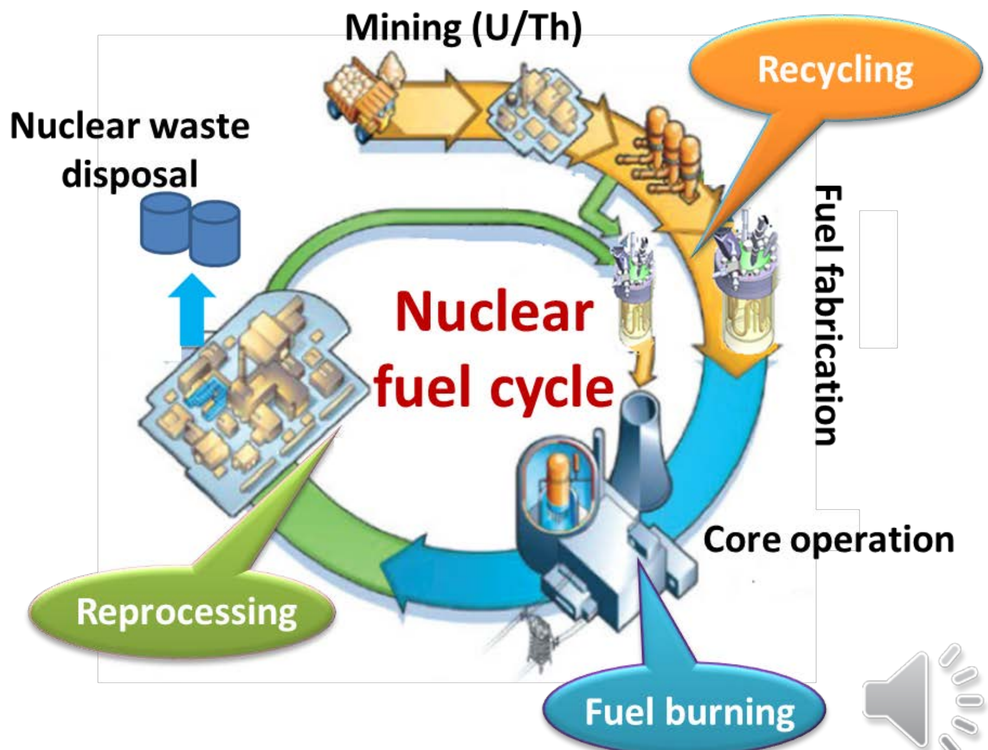
- Discovering fundamental physical laws of Th-U fuel cycle in MSRs
- Created a reprocessing flow sheet and demonstrated it in cold, lab-scale facilities
- Established a comprehensive evaluation method assessing Th-U fuel cycle

– Th-U fuel cycle mode and thorium utilization scheme in MSRs

– Physical laws and chemical behaviors of actinide elements in the core

– Flow sheet of reprocessing and key technologies development

– Comprehensive evaluation (economy, safety, non-proliferation, et al.) of Th-U fuel cycle



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Research progress of TMSR Th-U fuel cycle

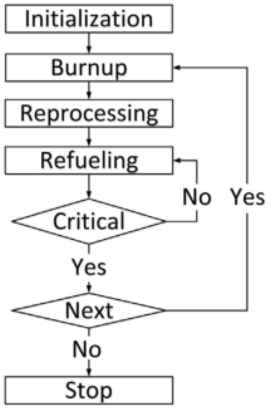
--Fuel cycle performance & physical laws

Development of MSR burnup simulation tools

Realized MSR reprocessing and point-depletion burnup simulation, performed Th-U cycle transition and TRU transmutation analysis

R&D of MSR fuel cycle simulation tool

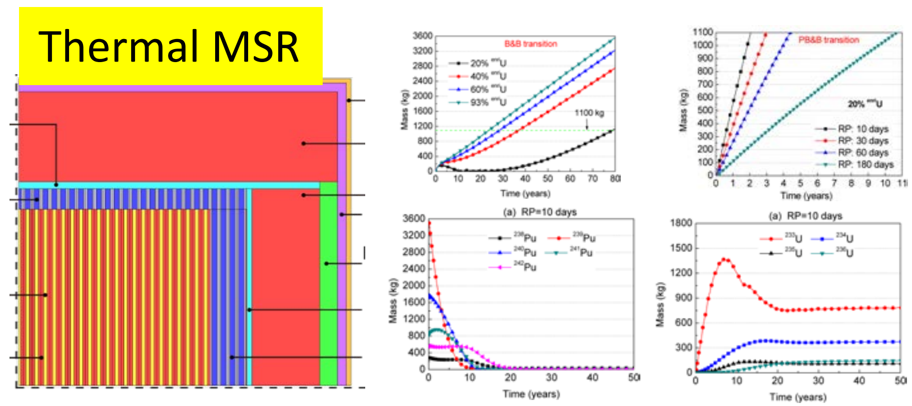
- **MSR-RS (MSR reprocessing sequence) code:** realizing MSR on-line refueling, batch/on-line fuel reprocessing simulation



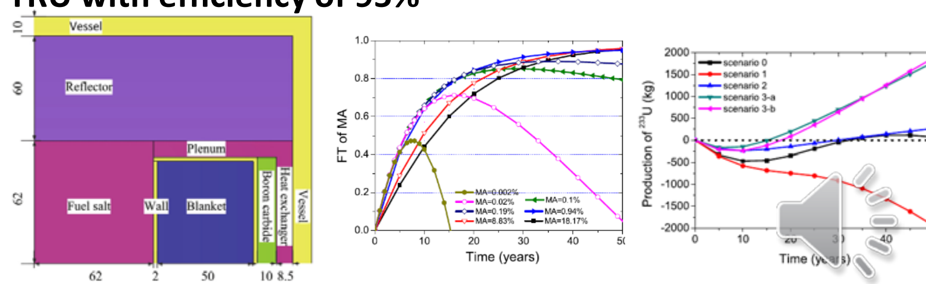
- **MSR point-depletion code:** using newly accuracy depletion method (Chebyshev rational approximation method, CRAM) to solve the burnup equations with a continuous feed, validated with ORIGEN-S

Conducting analysis of transition towards Th fuel cycle & TRU transmutation in MSR systems

Enriched U and Pu from LWR can realize transition towards Th fuel cycle with B&B and PB&B scenarios



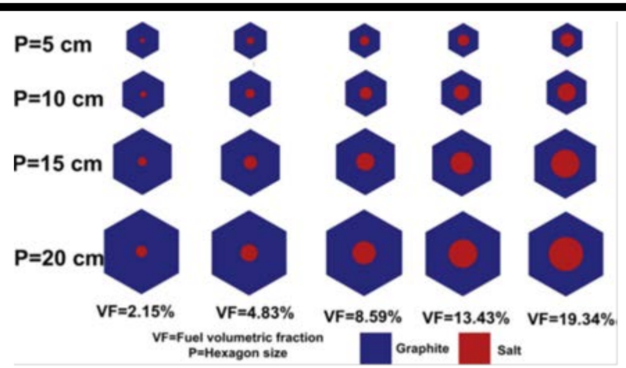
Thorium Molten Salt Fast Reactor can effectively transmulate TRU with efficiency of 95%



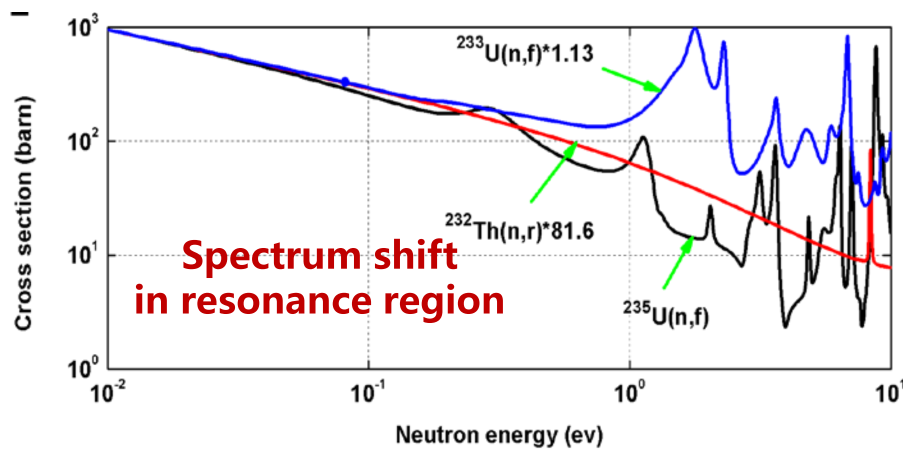
Performance of Th-U fuel cycle in MSRs

Research on Th-U breeding in a thermal MSR

- Realizing Th-U breeding with a negative temperature feedback coefficient
- Key problem： large positive temperature coefficient in graphite



Optimization on graphite assembly in thermal spectrum range and select an optimal value



Cross-section competition of U3, U5 and Th2 on the whole energy range

Nuclear Engineering and Design 281 (2015) 114

Progress in Nuclear Energy 104 (2018) 75

core	Reprocessing period	Reprocessing ability	Breeding ratio	Doubling time
TMSR	10d	1000L/d	1.045	13 y
MSBR	10d	4620L/d	1.038	31 y
MSFR	450d	40L/d	1.11	56 y

- A negative temperature coefficient in a thermal MSR is occurred due to a spectral shift propitious to U-233 fission
- Thorium breeding is achieved in a thermal MSR by optimizing geometry of graphite cell

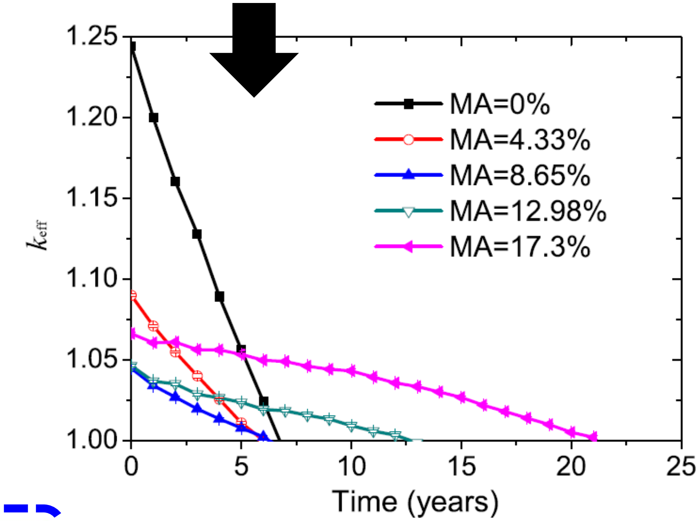
Th-U breeding and TRU transmutation in a MSFR

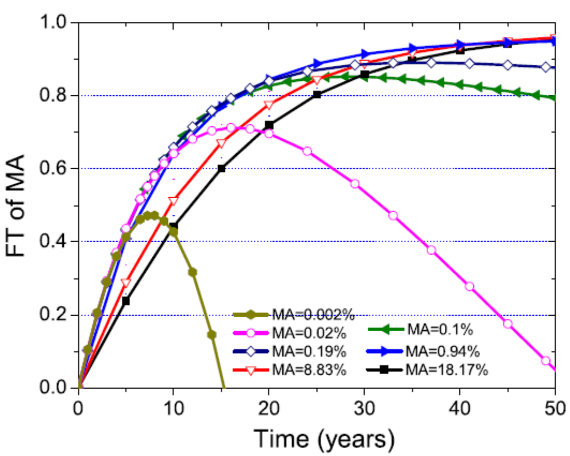
- **Objective:** **High TRU transmutation within Th-U iso-breeding**
- **Key issues:**
  - Solubility of TRU in molten salt limits its initial load and transmutation efficiency.
  - Accumulation of U-233 in the fertile salt affects the Th-U breeding capacity.

Annals of Nuclear Energy 85 (2015) 597

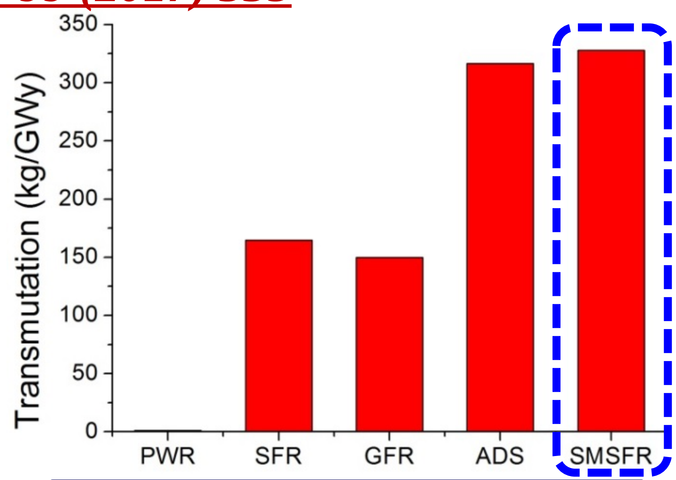
Annals of Nuclear Energy 99 (2017) 335

optimization





TRU transmutation efficiency of 95%.



TRU transmutation capacity of 300 kg/GWthy

TRU transmutation in the Thorium Molten Salt Fast Reactor can solve the problems of nuclear waste disposal and nuclear fuel long-term supply

Transition for Th-fuel cycle in MSR

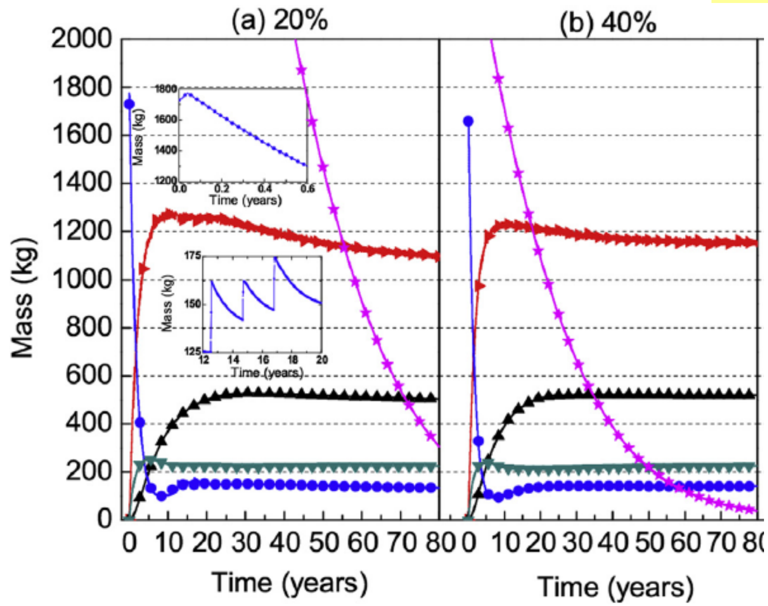
Goal: Producing ^{233}U for initial start-up;

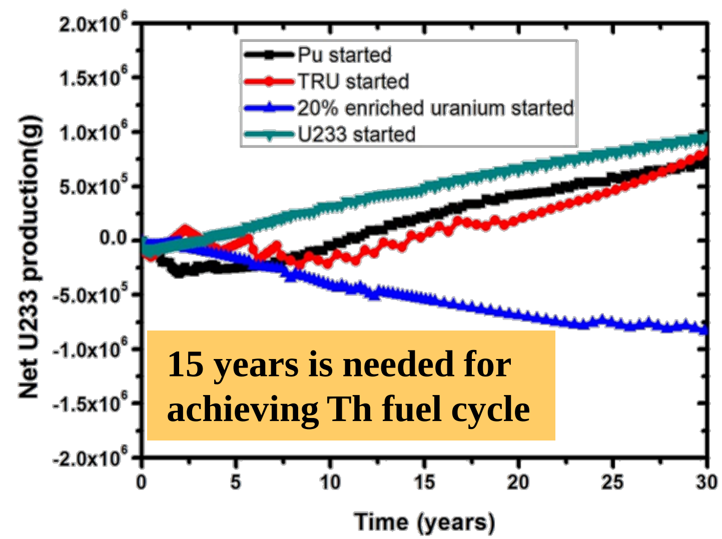
Problem: Transition methods and safety;

- **Absent ^{233}U in nature, obtained through ^{232}Th absorbing neutrons**

Initial fissile fuel

- ✓ **HEU**
- ✓ **TRU**
- ✓ **Pu**





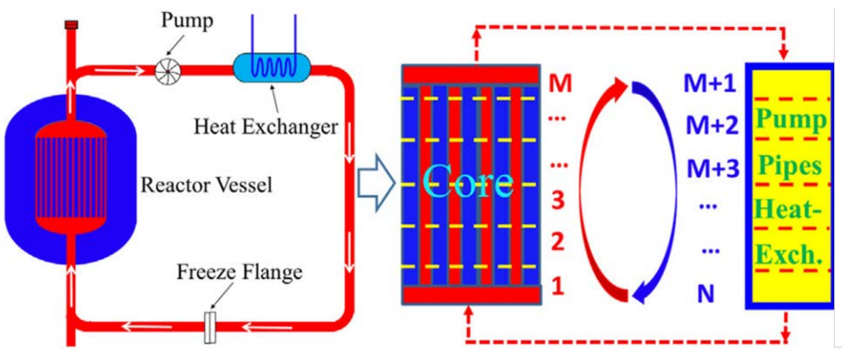
Nuclear Engineering and Design 339 (2018) 75

- For LEU (<20%), the fuel transition in core in difficult.
- Transition can be accomplished with Pu and TRU, the required time is about 15 years for B&B scenario and 5 year for pre-breeding scenario;

Influence of fuel salt circulation on nuclide evolution

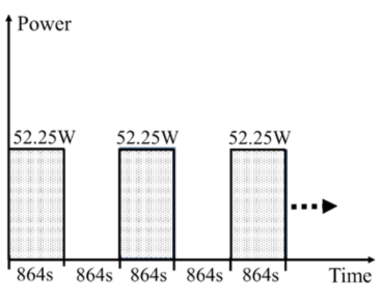
Code development and verification

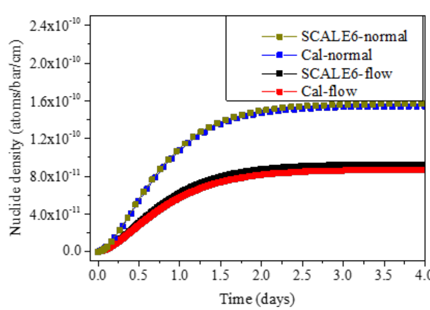
- Fuel salt flow modeling



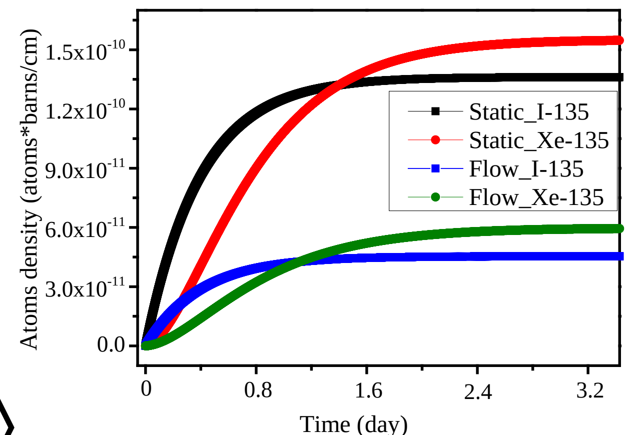
(a) reactor system (b) calculation model

- Code verification

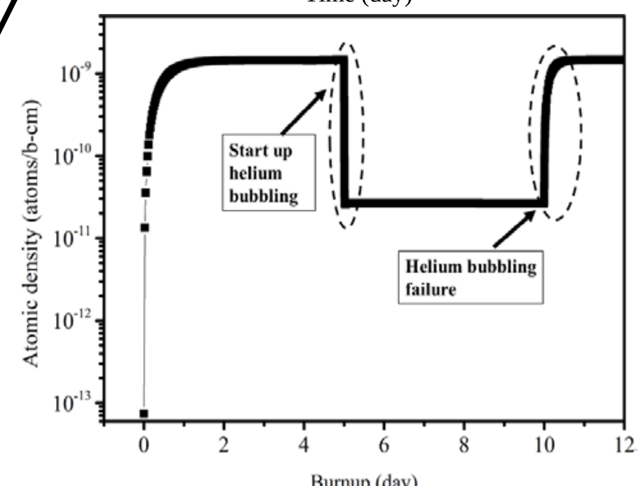




^{135}Xe concentration behavior



Fuel salt flow reduces nuclide concentration, increase reactivity ~700 pcm



Starting and losing Helium bubbling function, Xe-135 greatly changed.

Nuclear Engineering and Design 314 :318–325

Annals of Nuclear Energy, 120, pp. 100-107.

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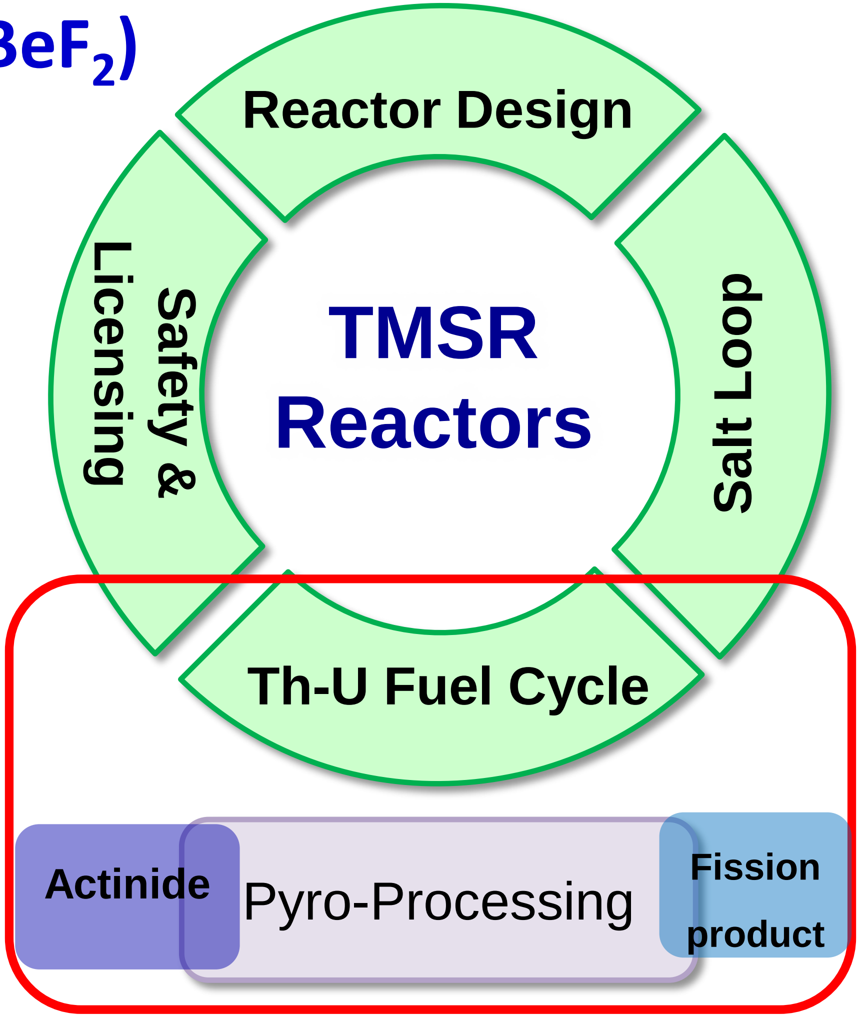
--Pyro-processing Techniques

liquid fluoride, (AnF_x-FPF_y-LiF-BeF_2)

- High radioactivity of fuel
- Poor water solubility of MSR fuel

Fuel Pyroprocessing

- Establish a reprocessing flow sheet
- Develop separation techniques
- Demonstrate in lab-scale facilities



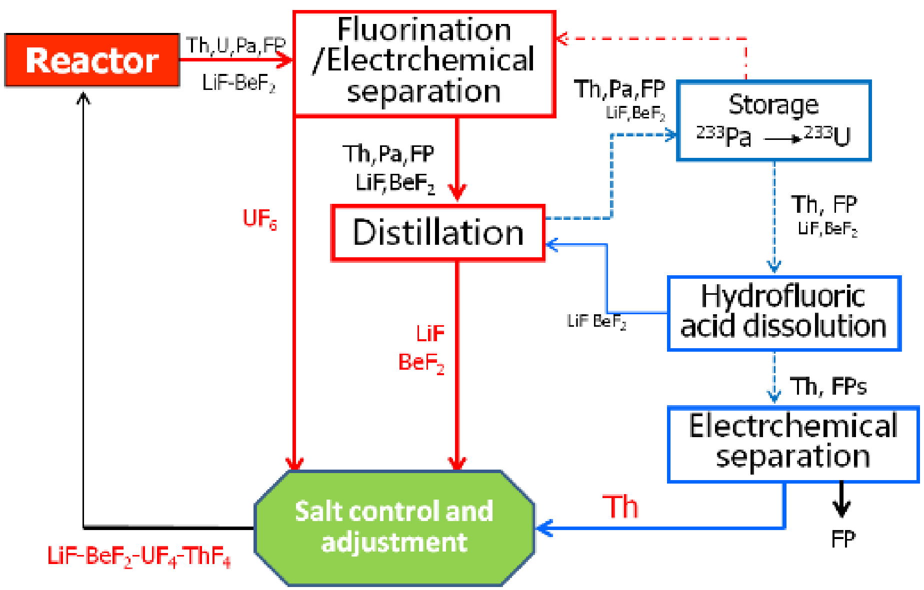
Flow Sheet of Th-U fuel cycle reprocessing

Flow sheet combines on-line and off-line processing

- On-line for U and carrier salt
- Off-line for Th and minor actinides

Consistency of on-line reprocessing in cold, lab-scale

- Recovery rate of U > 95%
- Recovery rate of carrier salt > 90%



Pyroprocessing techniques under development in SINAP

- Pyroprocessing techniques are under development on different scales in SINAP
- There are still fundamental issues that have to be understood during the development of pyroprocessing techniques.

technique	target	principle	advantage
Fluoride volatility	U	•Low boiling point of product $UF_4+F_2=UF_6(g)$	•Matured technology •High recovery rate and purity
Low pressure distillation	$^7LiF-BeF_2$	•Different volatilities between molten salt and FPs	•No chemical reaction •Recovered salt can be recycled directly
Electrochemistry	Actinides	•Reductive deposition of metal on cathode $M^{n+}+ne^-=M$	•Relatively safe

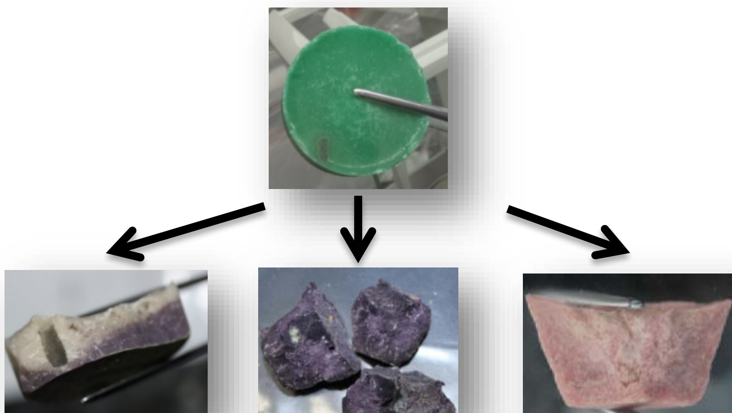
Influence of molten salt composition on the fluorination of UF_4

The fluorination of UF_4 in different molten salts

$UF_4(l) + F_2\ (g) \rightarrow UF_6\ (g)$

Before

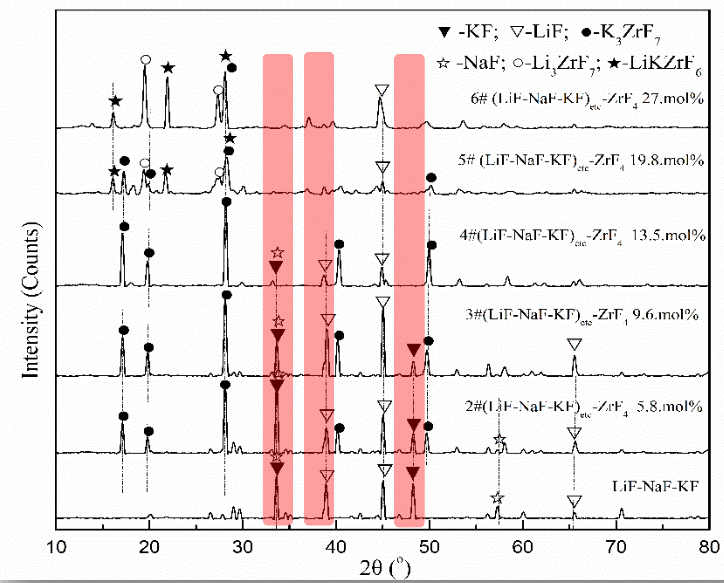
After



FLiNaK **FLiNaK-ThF_4(12mol.%)** **KF-ZrF_4**

The molten salts	U (wt%)		Fluorination time (min)	Conversion rate of UF_4 (%)	Average rate of fluorination (gUF_4/h)
	Before	After			
FLiNaK	1.37±0.07	1.35±0.10	120	0	0
FLiNaK-ThF_4	1.87±0.13	1.70±0.07	80	9.1	0.05
FKZr	1.46±0.12	42±9 ppm	50	99.7	0.69

Various amounts of ZrF_4 were added to FLiNaK to obtain the molten salts with different fluroacidities.

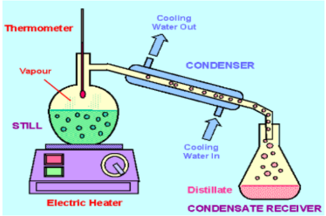


XRD spectra of FLiNaK with various contents of ZrF_4

Maybe, the difference in fluoroacidity was the major factor causing the difference of UF_4 fluorination behavior in the three molten salts.

Development of low Pressure Distillation techniques

A physical separation process occurred on the surface of a heating liquid



Vapor pressure difference

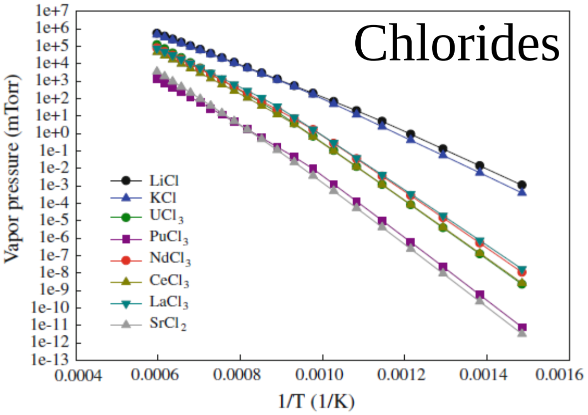
Advantages

- **Less by-products**
- **Less processing procedures**
- **Easier engineering amplification**

Achievement

- **Separate carrier salts from salt wastes.**
- **Minimize the high-level waste volumes.**

Volatility difference

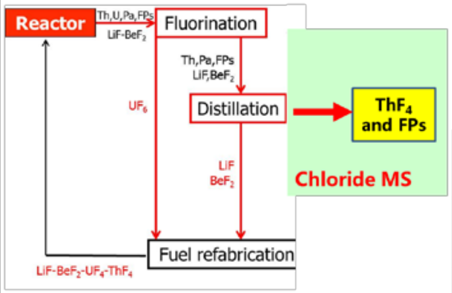


Fluorides

Molten salts	Vapor pressure / Pa	
	700 ºC	1000 ºC
FLiBe	＜10	1×10^3
FLiNaK	＜1	240
ThF_4	—	0.5
LnF_3	—	<0.01

- **Electro-separation of ThF_4**

Electro-separation of ThF_4 ---Change MS

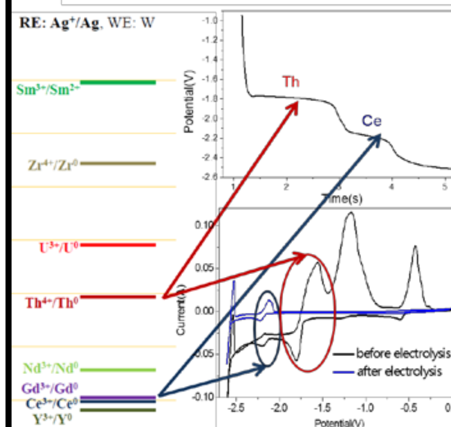


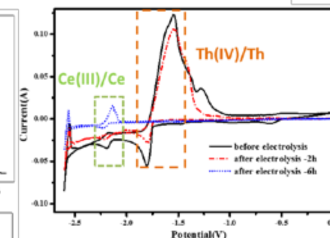
The reduction potentials of typical ions in molten salt ($LiF-BeF_2-ZrF_4$, 500℃, data from ORNL)

Ions couple	electrode potential (V)
Be^{2+}/Be^0	-2.12
Gd^{3+}/Gd^0	-1.97
U^{3+}/U^0	-1.74*
Zr^{4+}/Zr^0	-1.74
Cr^{2+}/Cr^0	-0.70
Fe^{2+}/Fe^0	-0.40

Th^{4+} can not be electrochemistry reduced in the $LiF-BeF_2$ MS

Electro-deposition of ThF_4 in Chloride MS





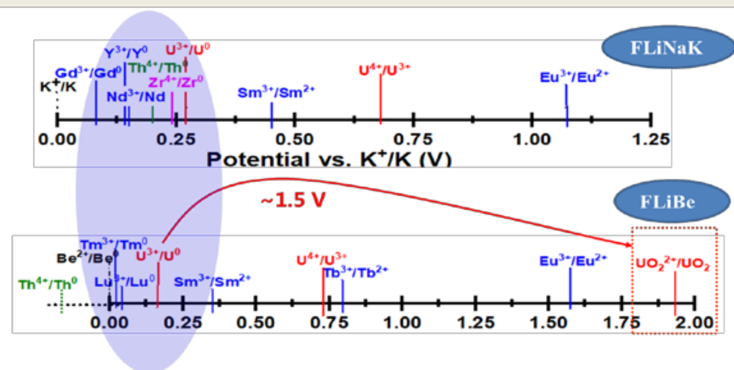
Recovery rate of Th: ~97% !

In MS	Before	After
Th (wt%)	1.21	0.036
Ce (wt%)	0.42	0.39

Electrochimica Acta 196 (2016) 286–293
Separation and Purification Technology 210 (2019) 236–241

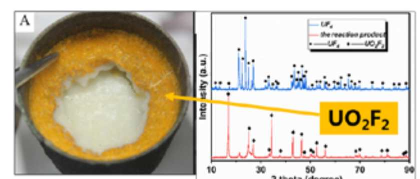
- **Electro-separation of UF4**

Direct deposition of UF_4 from REs in fluorides?

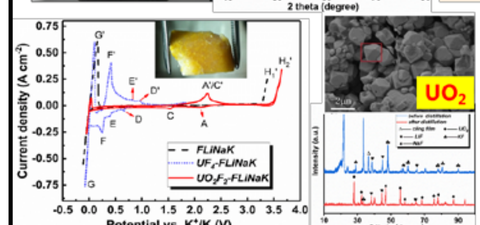


UF_4 → (Chemical transformation) → UO_2F_2 → (Cathodic reduction) → UO_2 + MS

New ideas of UF_4 electrochemical separation



- Transformation of UF_4 to UO_2^{2+} in fluorides
  $U^{4+} + O_2 \rightarrow UO_2^{2+}$
- Electro-reduction of UO_2^{2+} to UO_2
  $UO_2^{2+} + 2e^{-1} \rightarrow UO_2$



U separation in TMSR fuel

- $UF_4 \xrightarrow{F_2} UF_6$
- $UF_4 \xrightarrow{O_2} UO_2F_2$ ---- UO_2
- Mild reaction conditions

Journal of The Electrochemical Society, 165 (7) D301-D306 (2018)

OUTLINE

Motivation of TMSR Th-U fuel cycle

Advanced Th-U Fuel Cycle strategy

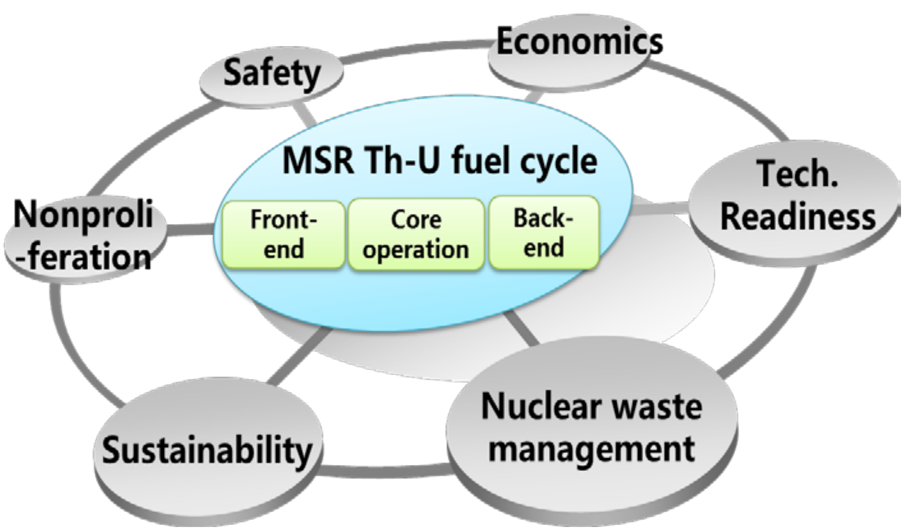
Research progress of TMSR Th-U fuel cycle

-- Fuel cycle comprehensive evaluation

Fuel cycle comprehensive evaluation have been studied worldwide

- Evaluation criteria: economy, safety, non-proliferation, sustainability, et al.
- A basis for judging whether the deployed nuclear system meeting the requirements of Gen-IV reactor system

1999	2000	2013	2014	2018
IAEA Qual. Eval.	**GIF** Qual. Eval.	**EPRI, USA** Qual. Eval.	**INL, USA** Semi-Quan.	**CNNC, China** Initiated



R&D in TMSR project

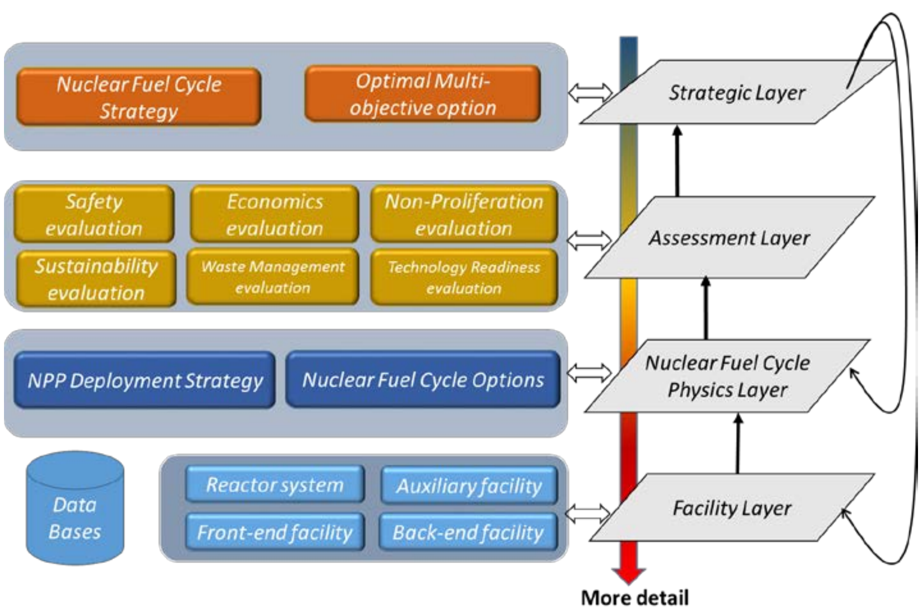
– Initiated in 2016
– Developed a quantitative evaluation method
– Evaluated the performance of TMSR “three-step” strategy

SINAP TMSR

Developed a quantitative evaluation method, TMSR-NUDEAS

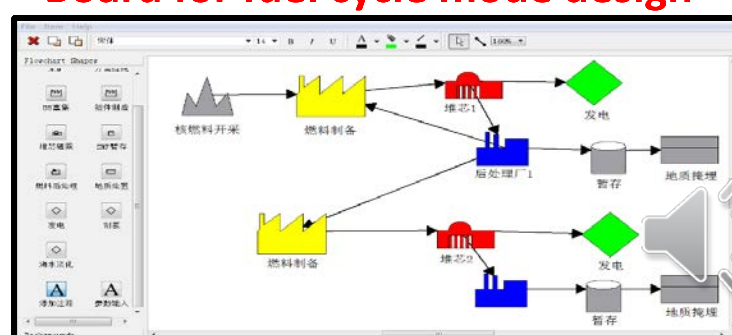
- Quantifying each aspect assessment by defining specified metrics and performing evaluation based on whole fuel cycle process
- TMSR-NUDEAS has a user-friendly interface, fuel cycle mode design board, displays results in real time

TMSR-NUDEAS （**TMSR NU**clear fuel cycle **D**ynamic **E**valuation and **A**nalysis **S**ystem）



Interface for parameters input

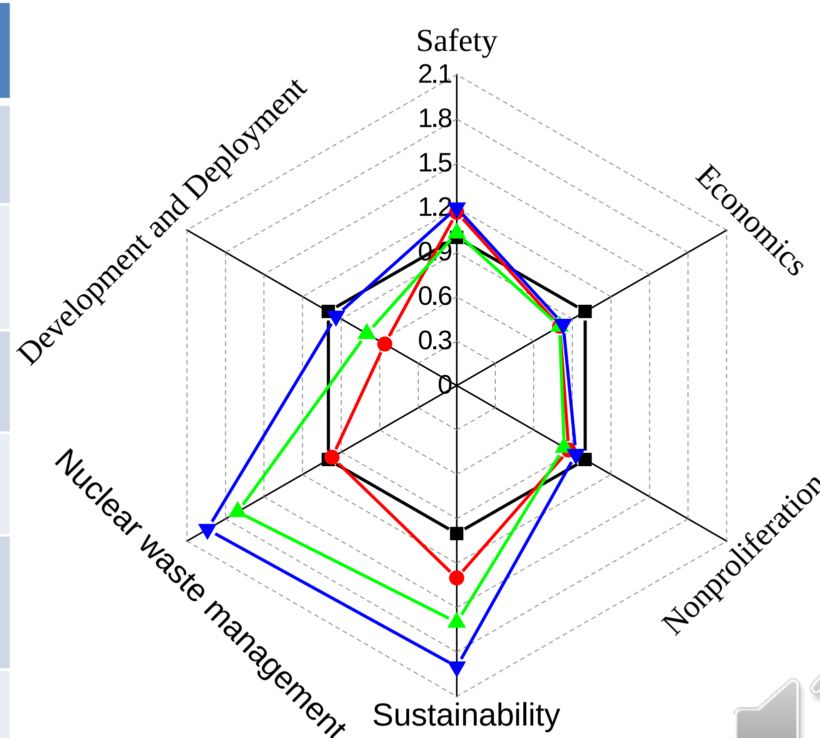
Board for fuel cycle mode design



Comprehensive evaluation of “three-step” strategy

- “Three-step” strategy presents a good performance relative to PWR, especially on sustainability and nuclear management, no water application and high burnup
- As steps go on, recycling Th&U in batch and improved technology readiness can further enhance the performance of fuel cycle

	Step 1	Step 2	Step 3
Safety	1.17	1.03	1.2
Economics **[KWh/$]**	20.3	21.5	21.03
Nonproliferation	0.79	0.69	0.78
Sustainability	3.25	3.75	4
Nuclear Waste Management	0.33	0.39	0.56
Development and Deployment	0.56	0.7	0.94



Nuclear non-proliferation overview, new method development

- Extensively reviewed the history, evaluation methods, et al. of non-proliferation
- Proposed a new quantitative evaluation method, MUT-PR (multi-theories proliferation resistant assessment method)

Investigated 150 related papers

Main issues

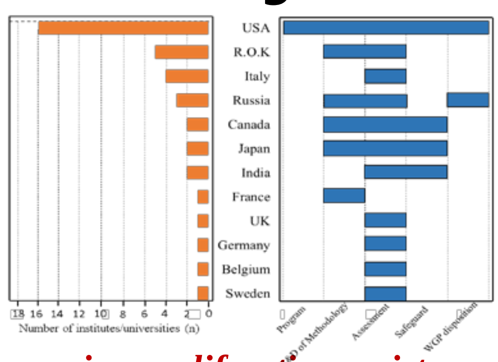
- Multi-process & parameters, hard to realize quantitative evaluation
- Large uncertainties
- Rare studies on influence of national policy on nonproliferation

Solutions

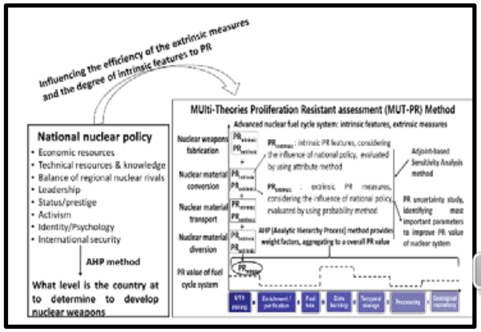
- Attribute method, probability method assess PR and PP respectively
- Adjoint-based sensitivity analysis method to handle uncertainties
- AHP method to assess influence of national policy

Non-proliferation

USA leading in studies

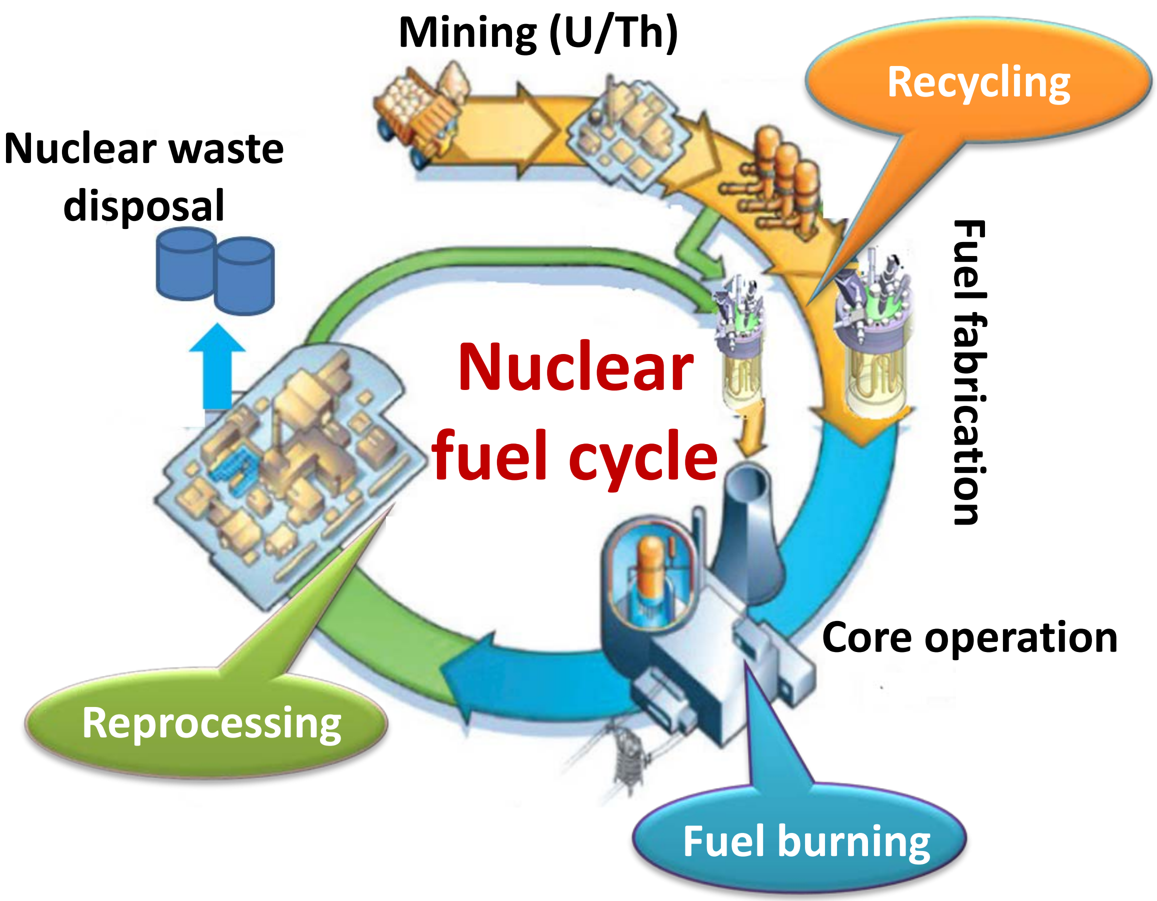


MUT-PR method



Nuclear non-proliferation review and improving proliferation resistance assessment in the future, 2020, Int. J. Eng. R.

Thank you for your attention



Mining (U/Th)
Recycling
Nuclear waste disposal
Fuel fabrication
Nuclear fuel cycle
Core operation
Reprocessing
Fuel burning