2014 International Pyroprocessing Research Conference (IPRC2014)
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Idaho National Laboratory, Idaho Falls, USA

Recent Progress on Development of the Pyrochemical Process of Spent Nitride Fuels for ADS*

* Accelerator-driven system

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Japan Atomic Energy Agency (JAEA)

R&D Activity on Pyrochemical Process in JAEA

Pyrochemical Reprocessing Development for Metal Fuel Fast Reactor Cycle

Advanced Fast Reactor Cycle System Research and Development Center, H. Kofuji, et al.

Department of Fukushima Technology Development, S. Kitawaki, A. Nakayoshi, et al.

CPF in Nuclear Fuel Cycle Engineering Laboratories, JAEA Tokai

Fundamental Research on Pyrochemical Process Including Treatment of Spent U-free Nitride Fuel for MA Transmutation using ADS

Nuclear Science and Engineering Directorate,

H. Hayashi, T. Sato, H. Shibata, M. Kurata, et al.

http://nsed.jaea.go.jp/en_index.html

NUCEF in Nuclear Science Research Institute, JAEA Tokai

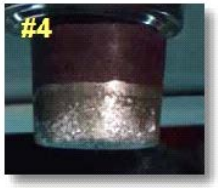
R&D has been promoted by the joint research with Central Research Institute of Electric Power Industry (CRIEPI)

Development of Pyrochemical Reprocessing of Metal Fuel

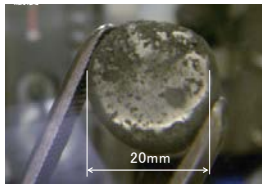
Japanese metal fuel cycle development for fast reactor is in progress as part of FaCT project. Pyrochemical reprocessing is being developed in collaboration with CRIEPI

◆ A series of experiments on assessing process efficiency with U-Pu-Am

Electrochemical reduction of MOX



Metal electrorefining in molten chloride



U-Pu-Am alloy ingot formation

◆ A novel approach to extracting TRUs from molten salt

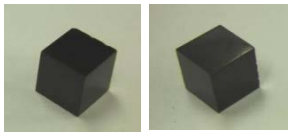
- Fundamental process tests have been conducted used U, Pu and Am in collaboration with CRIEPI
  ⇨Results of the study will be introduced in another session of this conference (Y. Sakamura, et al.)

◆ Other R&D for metal electrorefining process

◆ Concept study of the safeguard system for the pyrochemical reprocessing

- Conceptual design study
- Material accountancy technology

◆ Applicability study of iron phosphate glass for the reduction of high level waste volume



Glass sample

- Optimization of chemical composition
- Physical and chemical properties
- Dissolution test of FP elements

Situation of Radioactive Waste Management Study (1/2)

◆Concern to radioactive waste management has been increasing in Japan.

➔ **Transmutation technology** for long-lived nuclides (i.e. MA) is drawing the attention from public, media and politicians.

◆The Ministry of Education, Culture, Sports Science and Technology (MEXT) in Japan launched 2 working parties relating this issue (2012-2013).

- **Working Party on Monju Research Plan (FR)**
- **Working Party to review Partitioning and Transmutation Technology (mainly for ADS)**

◆Japanese Government issued the new "**Strategic Energy Plan**" in April, 2014.

Situation of Radioactive Waste Management Study (2/2)

Interim Report of WP for ADS (excerpted, informal translation)

Evaluation on R&D Issues and Future Plans

- The evaluation of technical readiness levels of the technical fields (**partitioning, ADS, fuel cycle, and fuel**) showed that we are approximately on the stage to move up R&D from "conceptual study" to "principle demonstration", **and it is appropriate to shift the R&D to the next stage of engineering scale**.

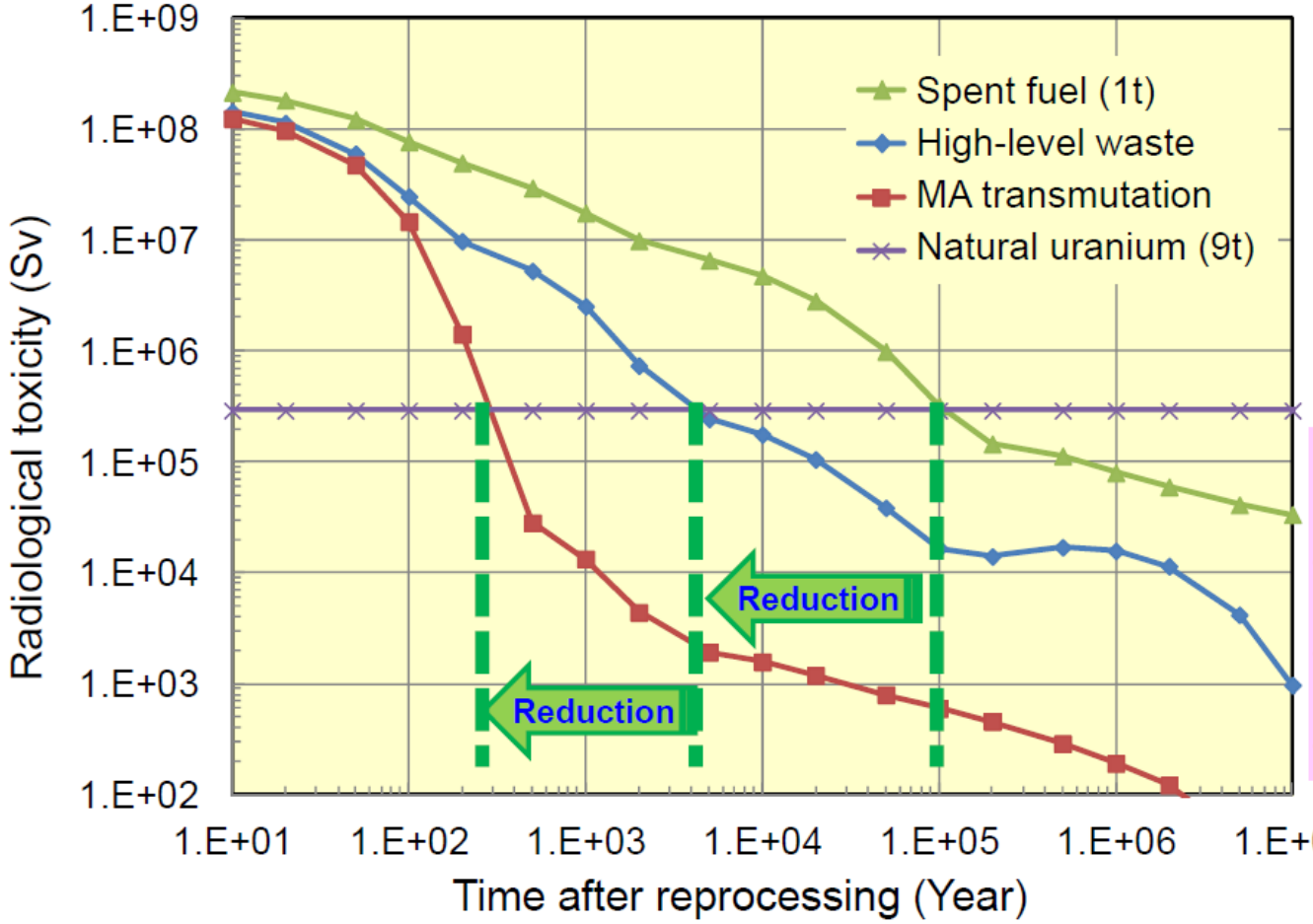
Roadmap

- It is important to promote the R&D of P-T technology with keeping consistency among various fields of technology. (snip) **The roadmap proposed by JAEA is regarded as appropriate**.

H. Oigawa (2014), "Review of ADS and P&T Program in Japan", OECD/NEA 13th Information Exchange Meeting on PT Seoul, Republic of Korea, September 23-26, 2014

Purpose of the MA Transmutation

Reducing the burden of the nuclear waste disposal



- Normalized by 1t of spent fuel.
- 9t of natural uranium (NU) is raw material of 1t of low-enriched uranium including daughter nuclides.

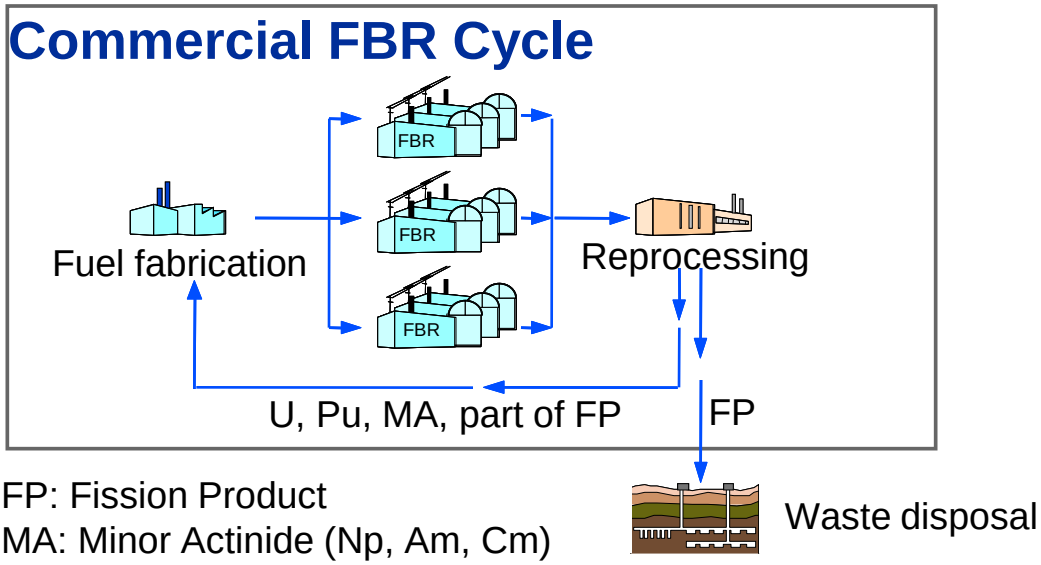
Time period to decay below the NU level:

- Spent fuel 100,000y
- ↓
- High-level waste 5,000y
- ↓
- MA transmutation 300y

H. Oigawa (2013), "ADS Study in JAEA", International Symposium on Nuclear Back-end Issues and the Role of Nuclear Transmutation Technology after the accident of TEPCO's Fukushima Daiichi Nuclear Power Stations, Kyoto, Japan, Nov. 28, 2013.

Homogeneous Recycle in FBR

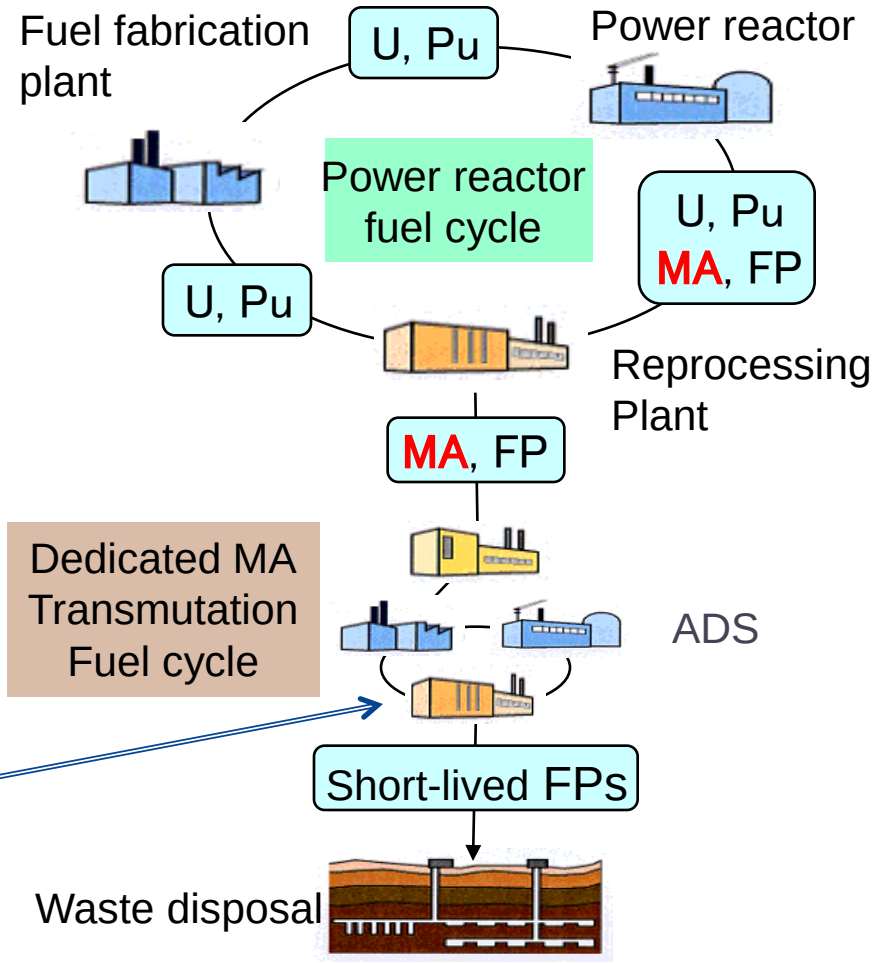
- MA is transmuted by commercial FBR power plants
- 5% MA (max.) is added to conventional FBR fuel (MOX, Metal, ...)



FP: Fission Product
MA: Minor Actinide (Np, Am, Cm)

Dedicated Transmutation by ADS (Double Strata Concept)

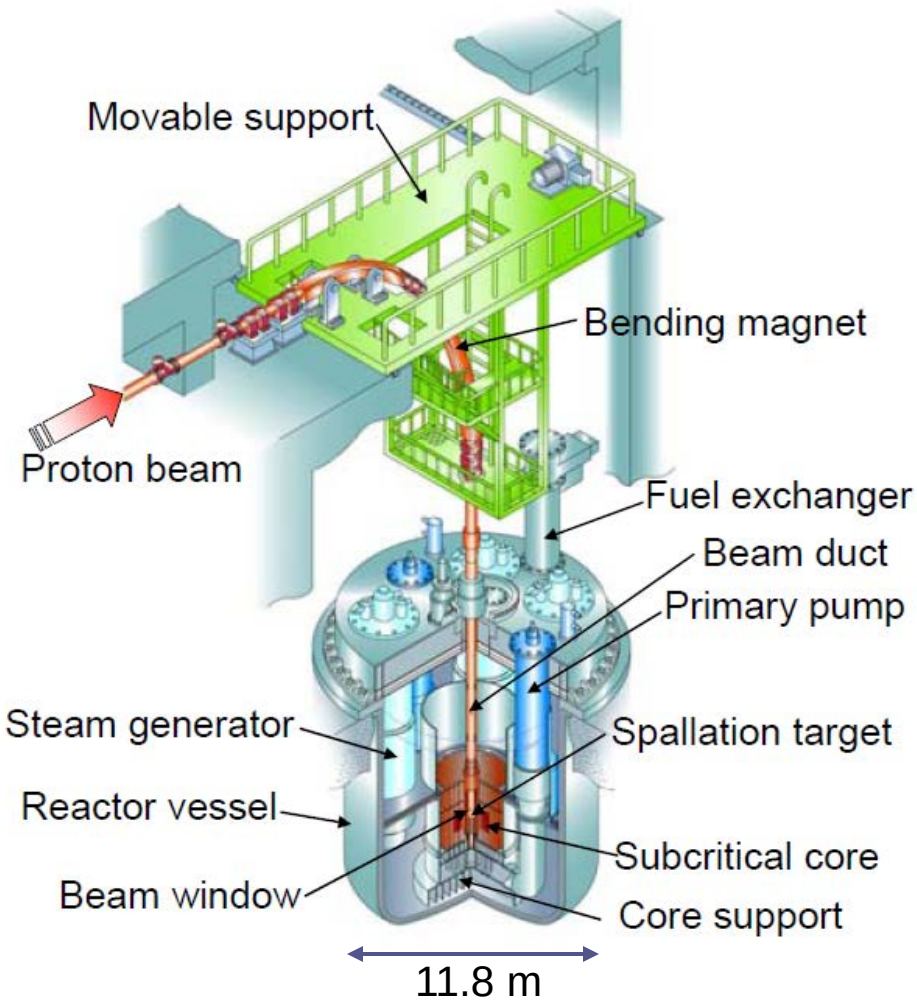
- Each fuel cycle pursues safety and economy of the fuel cycle independently.
- Performance of the power reactor fuel cycle is not disturbed by MA.
- Hazardous MA are confined in the dedicated MA transmutation fuel cycle with a small throughput.



- **Pyrochemical process is proposed for the treatment of spent nitride fuel for ADS***

* Accelerator-driven system

Design Study of ADS and MA Nitride Fuel



Conceptual view of 800 MWth LBE-cooled ADS

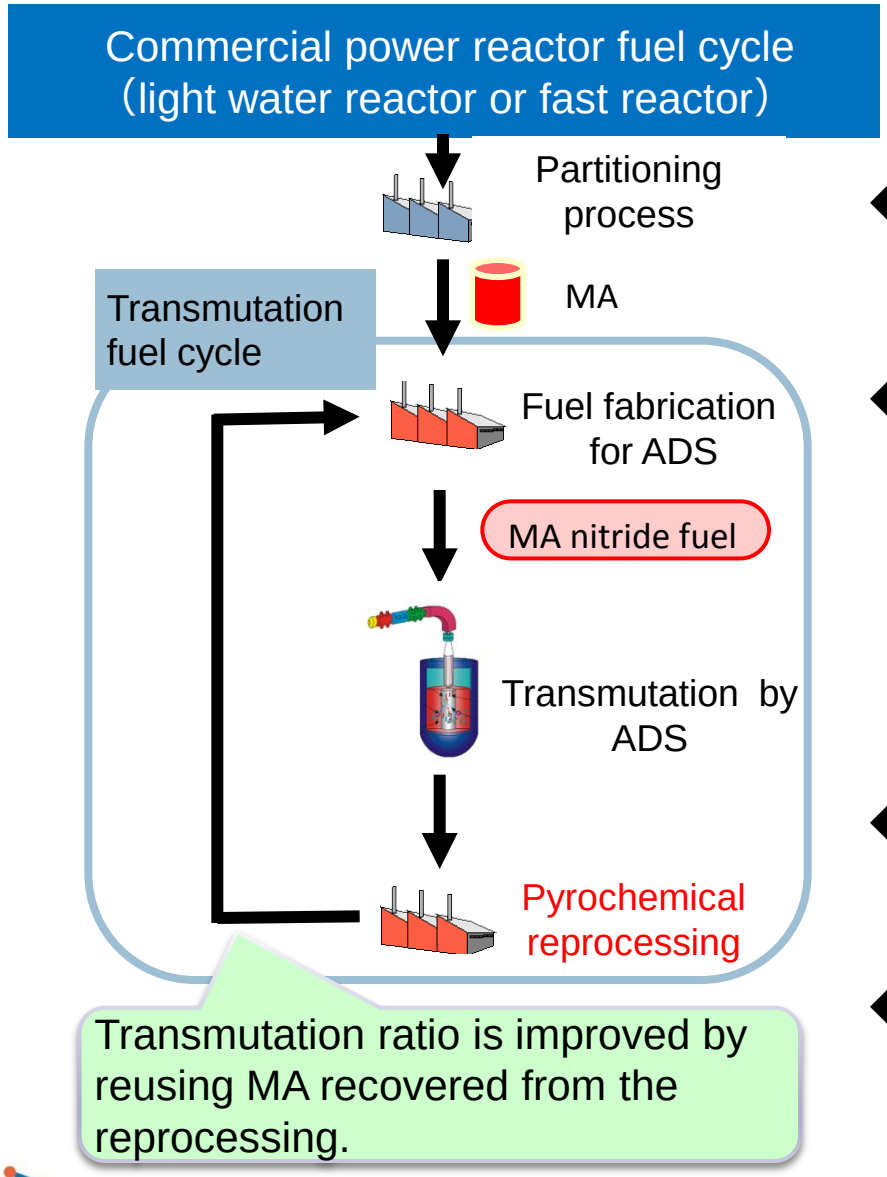
Basic design parameters of ADS

Parameters	Specification
Thermal power	800 MW
Electric power	270 MW
Coolant (average inlet / outlet temperature)	LBE (300 °C / 407 °C)
Spallation target	LBE (window-type)
Accelerator	Superconducting LINAC
Proton beam energy	1.5 GeV
Active core diameter	234 cm
Active core height	100 cm
Fuel	(MA, Pu)N
Inert matrix	ZrN
Effective multiplication factor (k_{eff})	Max. : 0.970
Cycle length	600 EFPD
Transmutation rate	500 kgMA/cycle

・MA nitride fuel for ADS

- MA is contained as a principal component in the fuel
- Diluent material is contained in place of U
- Highly enriched ^{15}N is used to avoid ^{14}C formation.

MA Transmutation Fuel Cycle



- The total inventory in an ADS core is Pu+MA: 4400kg（MA2640kg).
- 500 kg of MA* is transmuted during each burn-up cycle (600 effective full power days (EFPD), 800 MWth power).

  *500 kg of MA ~ produced in 10 LWR in 2 years.
- The transmutation ratio of MA per cycle is expected ～20%.
- All the fuel used in each cycle are removed from the core and Pu+MA remained in the spent fuel is recycled for the next burn-up cycle.

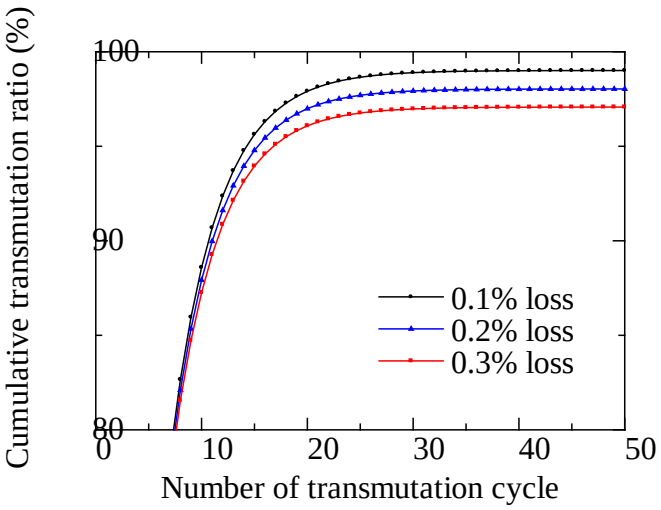
Achievable MA Transmutation Ratio

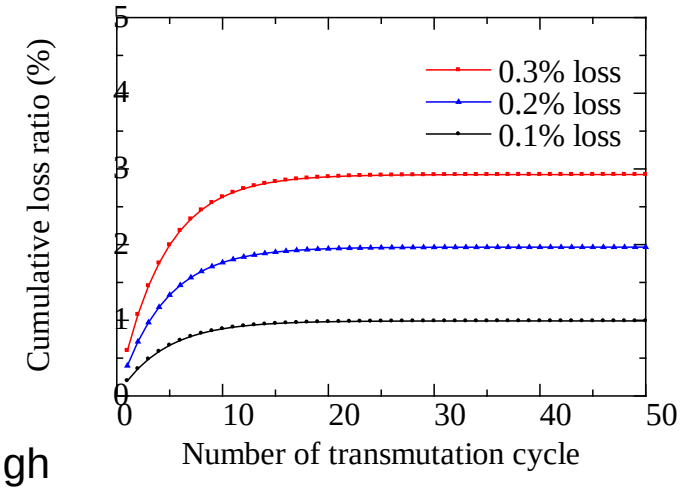
Our target is to transmute 99% of MA arisen from high level waste (HLW).

- The cumulative transmutation ratio & the cumulative loss of MA elements vs number of the cycle are calculated with
  - Transmutation ratio per cycle = 20%
  - The ratio of the loss on each process*

  = 0.1, 0.2, 0.3 %.

  *partitioning, fuel fabrication, fuel reprocessing





- Transmutation ratio per cycle > 20%, and
- The ratio of the loss on each process < 0.1% (recovery ratio > 99.9%) are necessary to transmute 99% of MA .

Development of the technology suitable for high recovery ratio is needed.

Nitride Fuel Fabrication Process

HLW from commercial power reactor cycle

Partitioning Process

Pyrochemical treatment

MA containing aqueous solution

Recovered TRU (TRU-Cd alloy)

Carbothermic reduction

via oxide

Nitridation-distillation combined method

MA nitride

TRU nitride

Inert matrix(ZrN)

Powder metallurgy

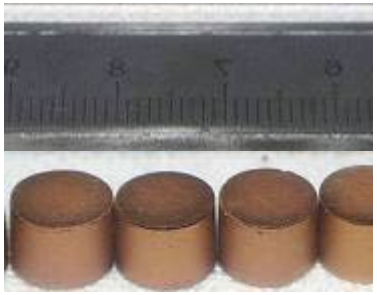
Inert matrix nitride fuel powder material

Nitride fuel pellet

Fuel rod

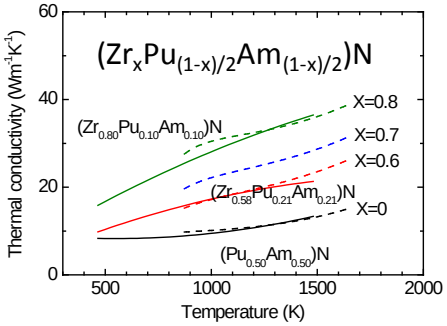
Fuel assembly

(Zr,Pu,Am)N disk sample 3mmϕ、95mg

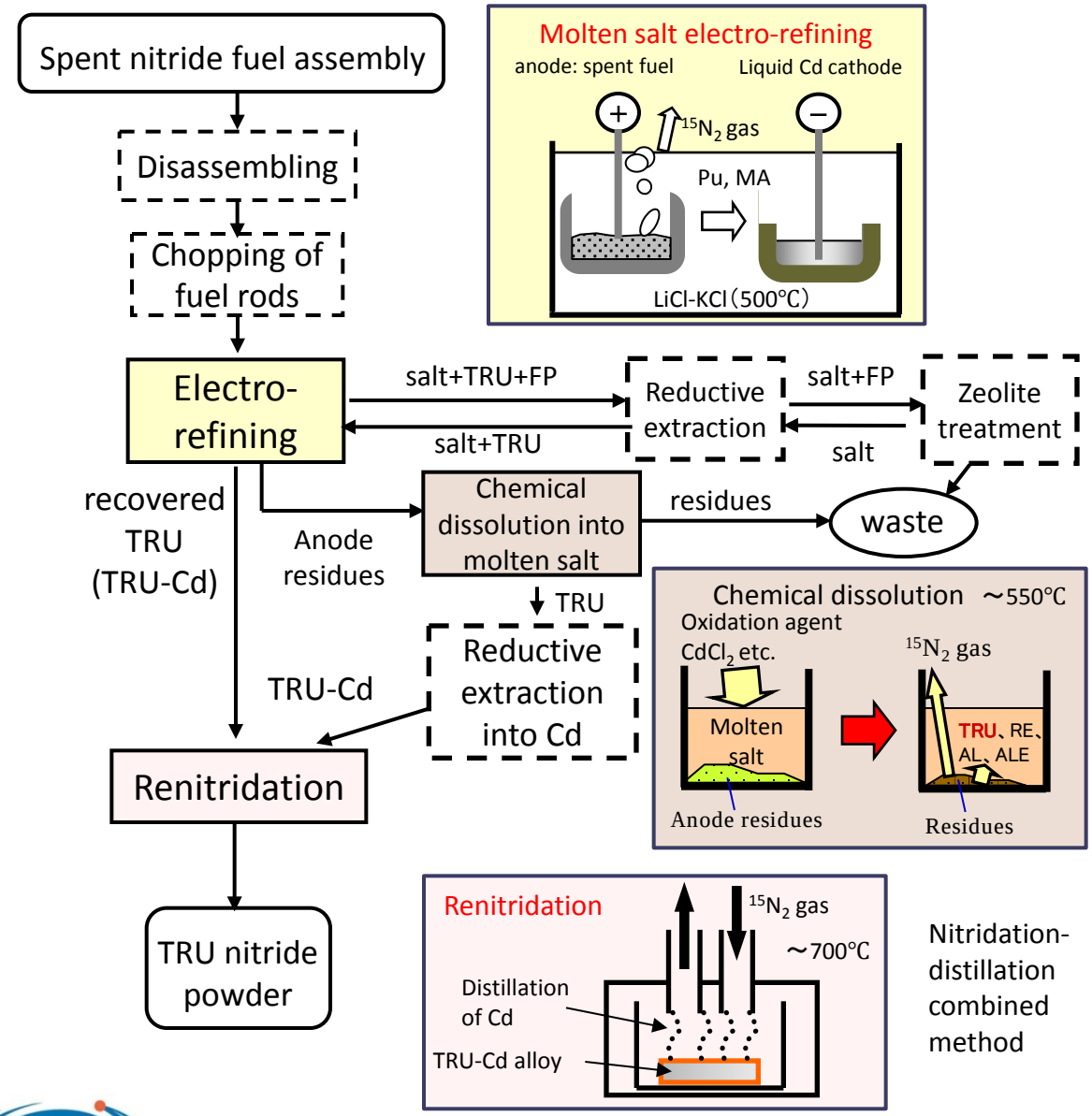


(Zr,Pu)N pellet sample for irradiation test at JMTR

Property data of MA nitride fuels were compiled in the databook (JAEA-Data/Code 2014-001).



Pyrochemical Process of Spent Nitride Fuel



- The process flow is based on that of metal fuel treatment.
- Electro-refining is applicable because of high electric conductivity & relatively low stability of nitrides.
- Generation of ^{15}N gas with dissolution of MA into molten salts occurs.

- Spent fuel from 1 ADS plant contains 3.9 t of TRU (Pu+MA).
- The amount of TRU handled in the process: 3.9t / 2years = 10 kg/day (20 kg spent fuel/day).

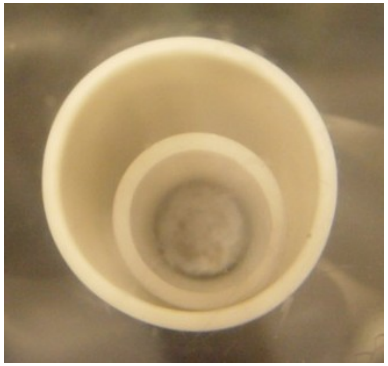
A Series of Experiments for (U,Pu)N

(U,Pu)N pellet
in Mo crucible

Electro-
refining

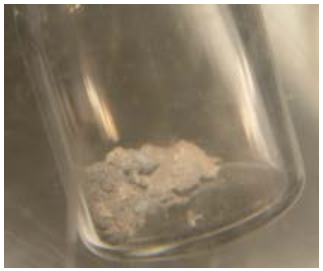
Liquid Cd cathode
in Al_2O_3 crucible

Cutting



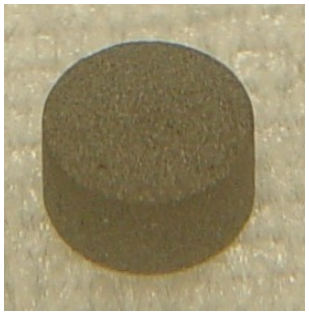
U-Pu-Cd alloy in
Y_2O_3 crucible

Renitridation

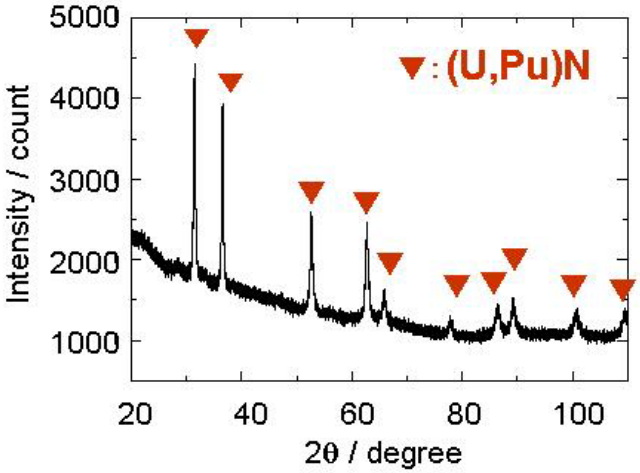


(U,Pu)N powder

Sintering

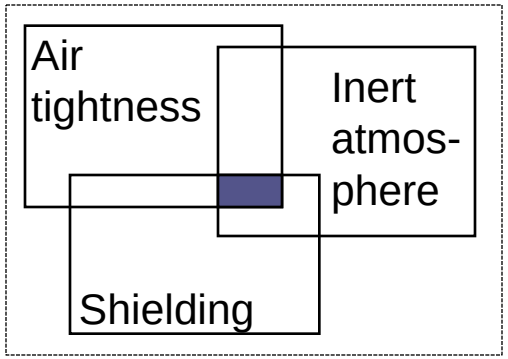


(U,Pu)N pellet



- ✓ Single phase of (U,Pu)N
- ✓ Density: ~84%TD
- ✓ O_2 impurity: 0.1~0.2wt%

Experimental Study on Americium



The module for TRU High Temperature Chemistry, TRU-HITEC with high-purity Ar gas atmosphere installed in NUCEF facility.

Electrolysis of AmN

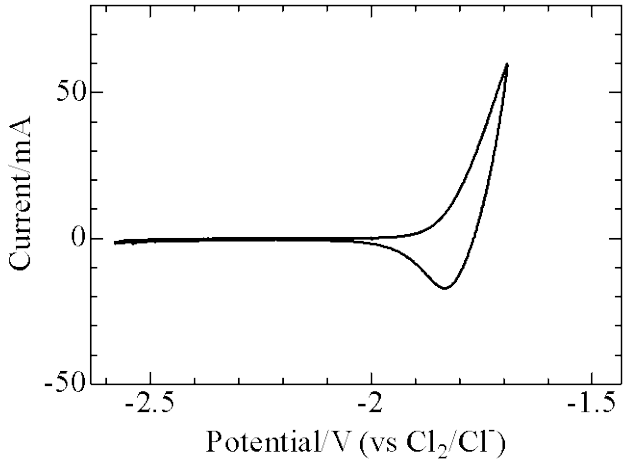
↓

Am recovered in Cd cathode (Am-Cd alloy) 28.9mg

973K,5h ↓ N_2 100cm^3/min

Fine black powder 10.4mg

Apparatus for electrochemical measurements



Cyclic voltammogram of AmN in LiCl-KCl eutectic melt at 723K

Fine black powder 7.2mg

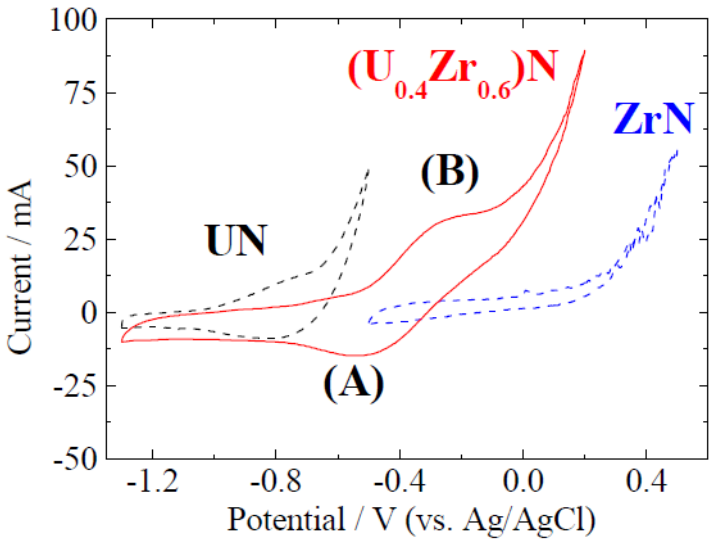
673K,3h ↓ in vacuum

to remove coexistent Cd

Heated sample 6.3mg

AmN powder

Electrolysis of Nitride Fuel Containing ZrN



- Addition of ZrN to nitride fuel
  - → Improving chemical stability
  - → Improving thermal conductivity
  - → Facing difficulty in reprocessing

Cyclic voltammograms for the dissolution of (U,Zr)N

Results of potential-controlled electrolyses of (An,Zr)N (An=U or Pu)

		$(U_{0.4}Zr_{0.6})N$		$(Pu_{0.1}Zr_{0.9})N$	
		U	Zr	Pu	Zr
Metal in initial sample (mg)		149	86	20	72
Metal in the salt (wt.%)	Initial	0.51	ND	1.10	ND
	Final	0.42	ND	1.09	ND
Metal left in the anode basket (%)		No sample left (dropped out)			
Metal recovered in the LCC (mg)		129	20	7.3	4.0
Recovery yield (%)		86	23	37	5.6

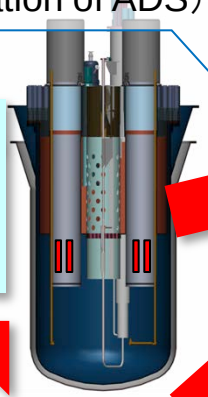
Actinides are selectively recovered in Cd cathode

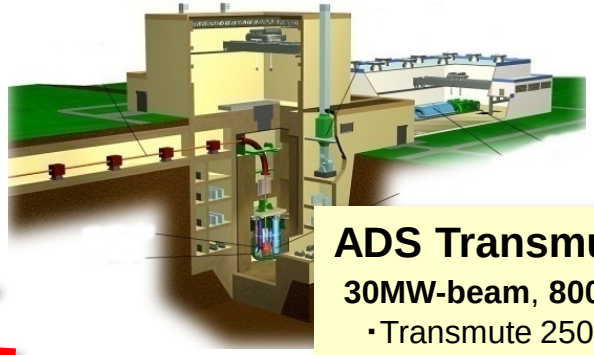
Further studies on the electrolyses of ZrN-containing nitride fuel using lanthanides are progress for proving the technological feasibility .

Development of ADS

Power

ADS without MA fuel
（LBE coolant, Accelerator, Operation of ADS）



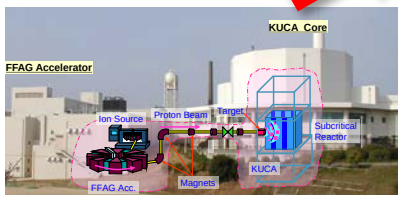


ADS Transmutation plant
30MW-beam, 800MW$_{th}$
・Transmute 250kg of MA annually

Experimental ADS➔MYRRHA
~2.4MW-beam, 50~100MW$_{th}$
・Engineering feasibility of ADS and fuel irradiation

Reactor physics of MA transmutation system and material development for spallation target material

Advanced material for beam window



TEF in J-PARC
250kW-beam
・LBE target technology
・Reactor physics of transmutation system

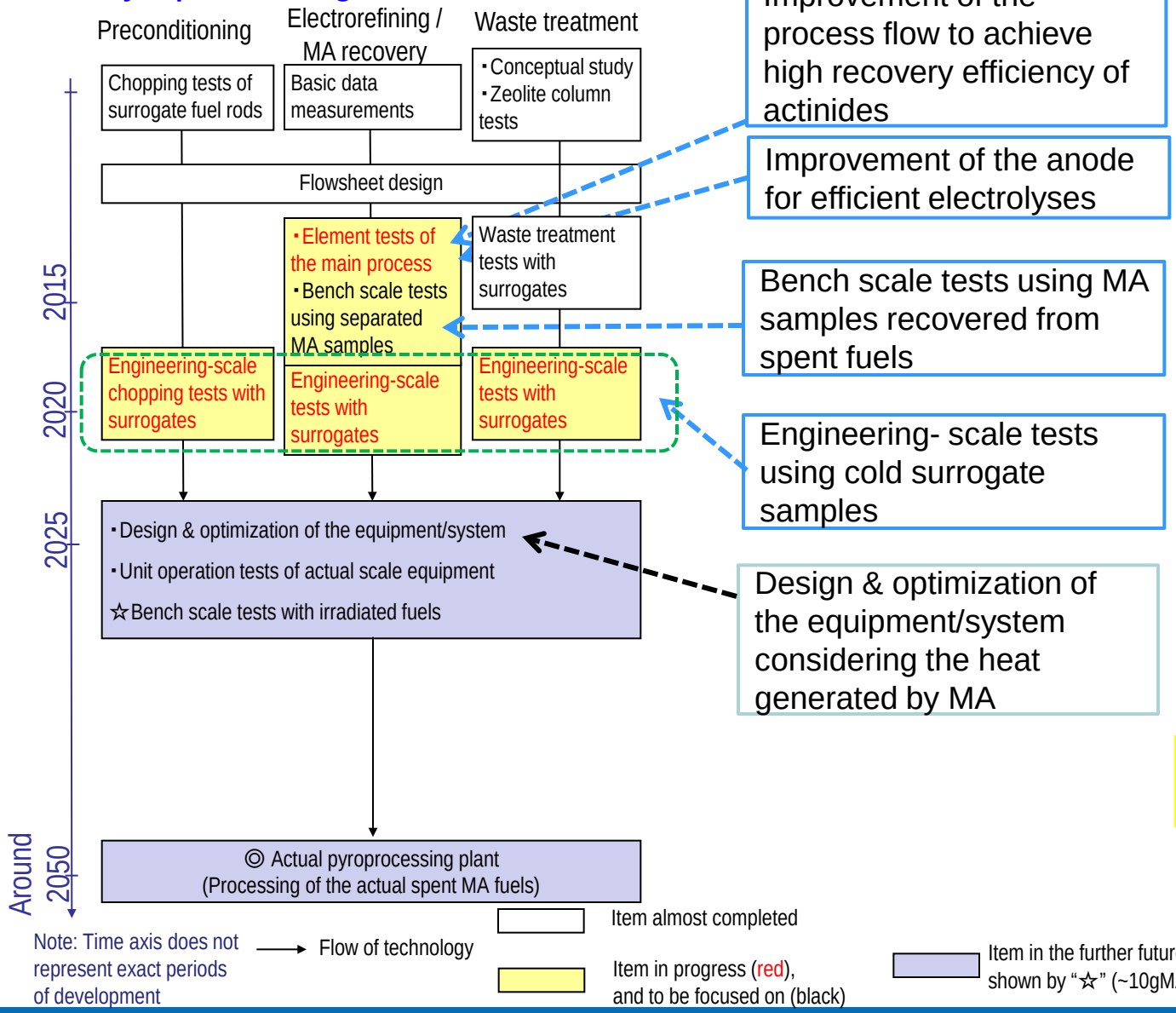
	TEF in J-PARC	MYRRHA
Purpose	R&D for elemental technology (LBE target, Reactor physics)	Fuel irradiation, Accumulation of operation experience of ADS
Power	250kW-beam TEF-P : 500W (max.)	2.4MW-beam Power : 50~100MW$_{th}$
MA	Mock-up experiment of transmutation system with massive MA (kg order)	Irradiation experiment with small amount of MA

Basic research (LBE loop test, KUCA experiments)

2010 2020 2030 year

Roadmap of the Development of Pyrochemical Process

Pyroprocessing of MA nitride fuel



Improvement of the process flow to achieve high recovery efficiency of actinides

Improvement of the anode for efficient electrolyses

Bench scale tests using MA samples recovered from spent fuels

Engineering- scale tests using cold surrogate samples

Design & optimization of the equipment/system considering the heat generated by MA

Utilizing the facilities and technology in JAEA and other organization in Japan

TRU-HITEC

Elecrorefiner for metal fuel

Domestic and global networks for cooperation

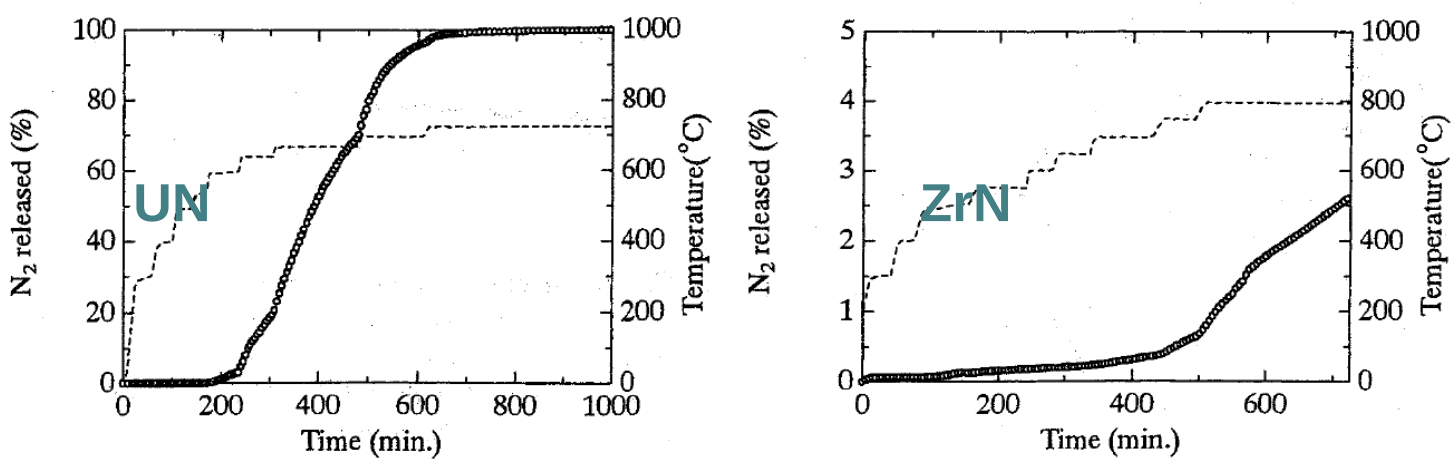
Summary

- Transmutation of long-lived nuclides (i.e. MA) is drawing attention in Japan.
- Using ADS is considered as one of the candidates for MA transmutation
- Technical feasibility of pyrochemical reprocessing technology for MA nitride fuel was demonstrated by laboratory-scale experiments.
- Basic design of the pyrochemical process flow is completed.
- Detail of the effects of containing ZrN in the fuel is being studied using surrogate materials.
- To prepare shifting the R&D to the next stage of engineering scale, roadmap of the development was determined.

Treatment of Inert Matrix Material ZrN

- The anode residue is considered to have ZrN as a main component.
- Removal of ZrN from AnN in the anode residue is possible by chemical process.
  - ✓ ZrN is more stable than actinide nitrides.

$$2AnN + 3CdCl_2 = 2AnCl_3 + 3Cd + N_2\uparrow$$



Dissolution behaviour of nitrides by the reaction with $CdCl_2$ in LiCl-KCl

"Strategic Energy Plan" issued in April, 2014

(excerpted, provisional translation)

http://www.enecho.meti.go.jp/en/category/others/basic_plan/pdf/4th_strategic_energy_plan.pdf

Chapter 3 Long-term measures regarding energy supply and demand to be implemented in a comprehensive and systematic manner

Section 4. Re-establishment of the nuclear energy policy

4. Steady approach without putting off implementing measures into the future

(1) Drastic reinforcement of measures for achieving solutions and promotion concerning spent fuel management

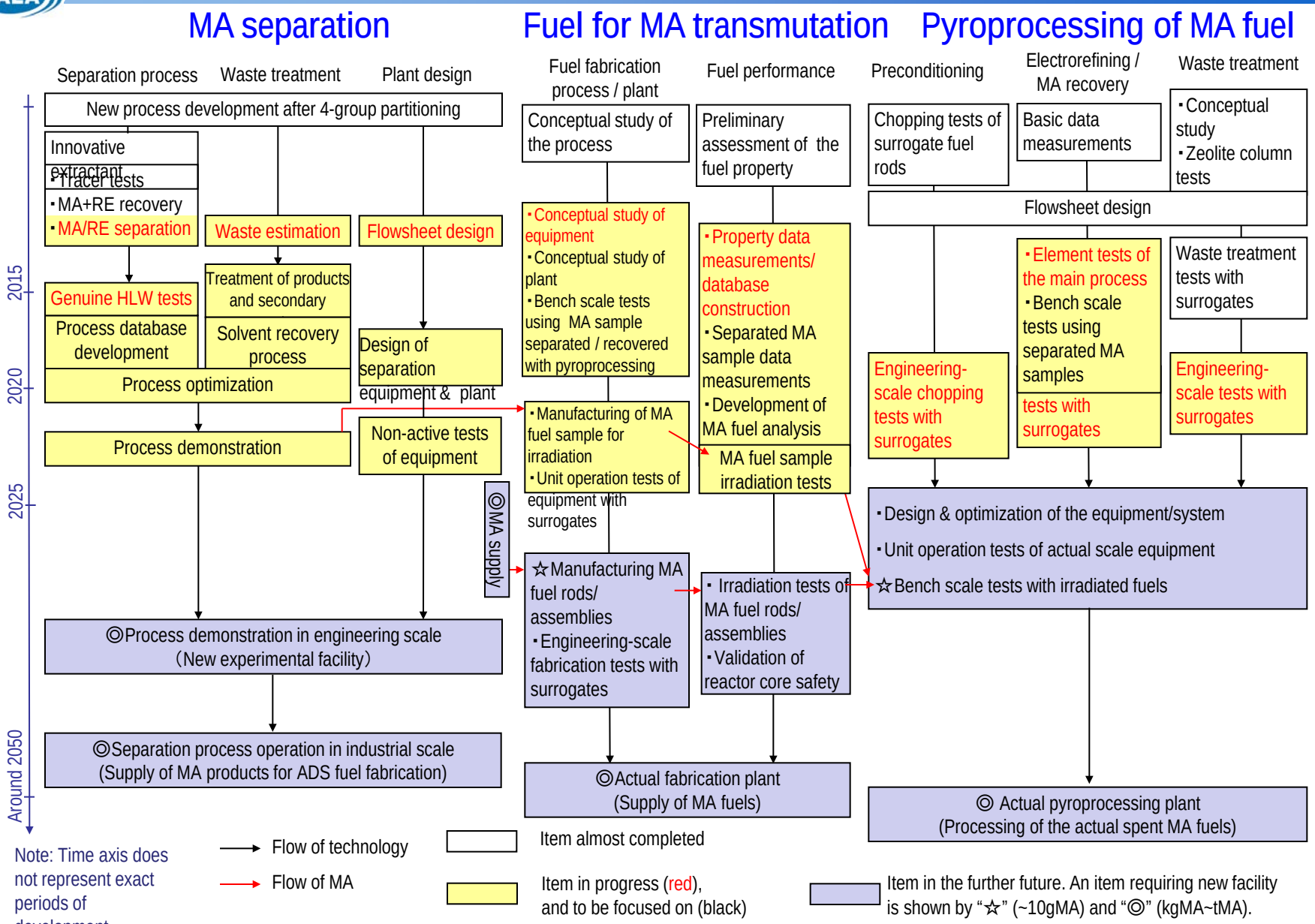
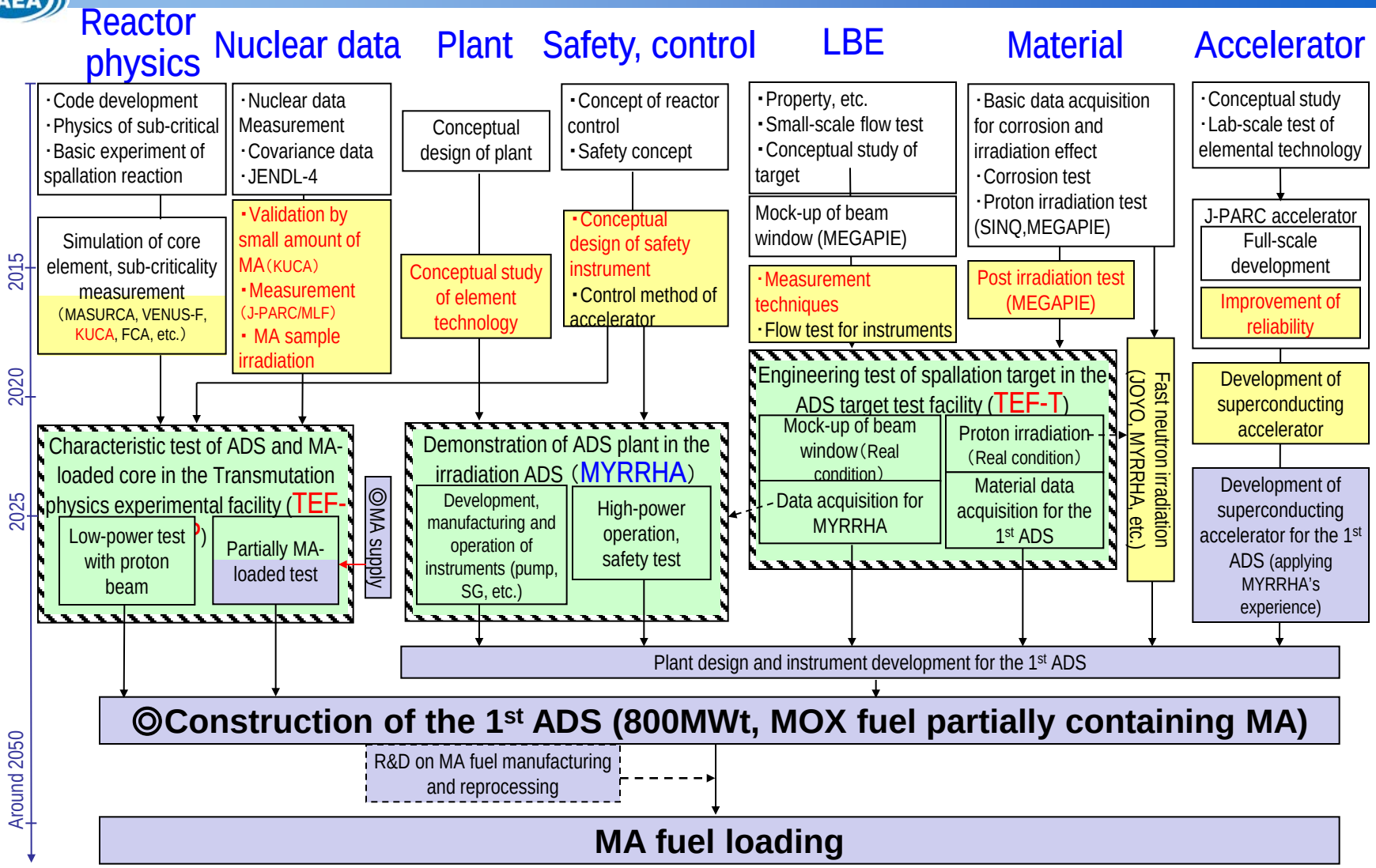
(iii) Promotion of technology development on volume reduction and mitigation of degree of harmfulness of radioactive waste

"Strategic Energy Plan" issued in April, 2014

(excerpted, provisional translation)

- GOJ will promote technology development on volume reduction and mitigation of degree of harmfulness of radioactive waste. Specifically, development of technologies for decreasing the radiation dose remaining in radioactive waste over a long period of time and enhancing the safety of processing and disposal of radioactive waste, including **nuclear transmutation technology using fast reactors and accelerators**, will be promoted by utilizing global networks for cooperation. Also, while GOJ examines the situation of study and progress in terms of final disposal, it studies the feasibility of integrated implementation of the R&D for final disposal and reduction of volume, international research cooperation and a researcher resource development related to them.

Overview of Future R&D plan: (1) Partitioning and ADS fuel cycle



Overview of Future R&D plan: (2) ADS plant