

# Report to the Sasakawa Peace Foundation

## Debris Treatment by Pyroprocess Technology

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(JFY2015 & 2016)**
- 2. Public literature search on Fukushima debris status**
- 3. Public literature search on debris treatment methods**
- 4. Pyroprocess for debris treatment**
- 5. Technical challenges**
- 6. Discussions**

# 1. Overview of the debris treatment study by pyroprocess

- Objectives
  - ✓ Evaluate technical possibility of treatment of the Fukushima Daiichi fuel debris by pyroprocess technology
  - ✓ Provide the preliminary R&D plan and the rough estimation of the cost/schedule for the demonstration facility
  - ✓ Identify technical challenges (R&D needs)
- Workscope
  - ✓ Public literature search on Fukushima debris characteristics/amounts
  - ✓ Public literature search on debris treatment methods
  - ✓ Evaluate promising pyroprocess methods for the debris treatment
  - ✓ Rough evaluation of the demonstration facility design and its cost/schedule
  - ✓ Identify technical challenges
  - ✓ Consideration on preferred metrics for nuclear waste transmutation from a viewpoint of public perception

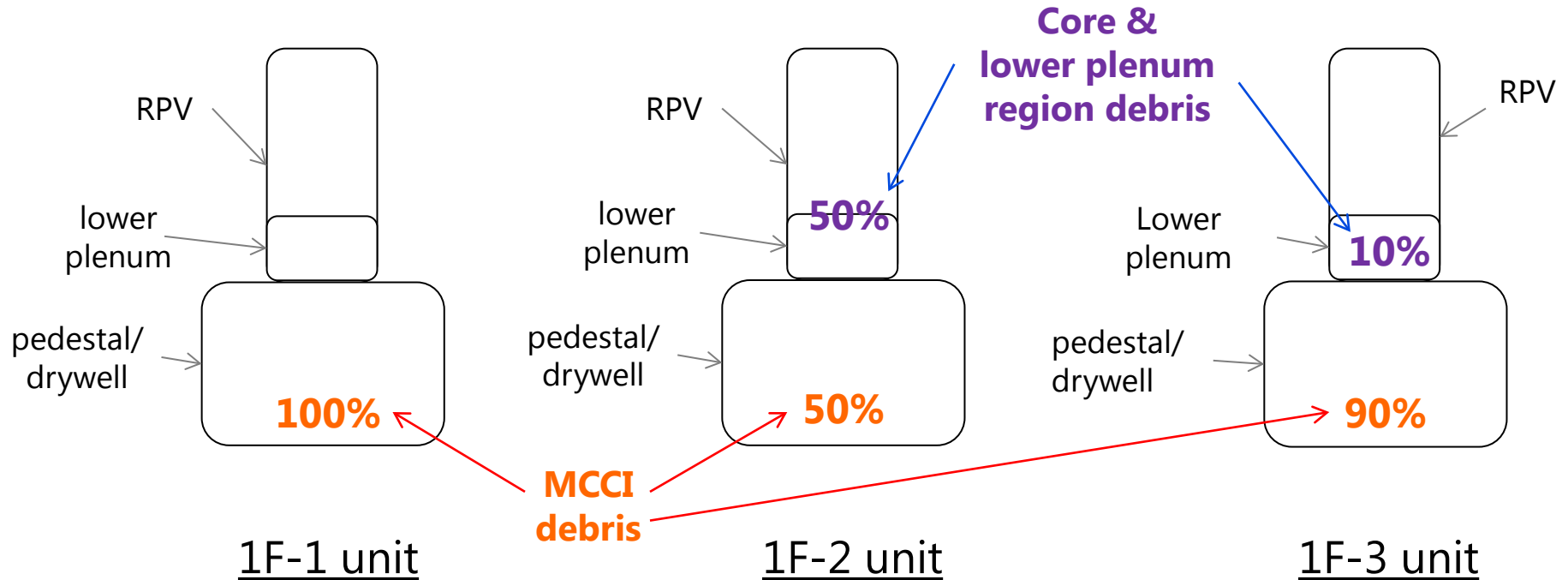
# 1. Overview of the debris treatment study by pyroprocess

## Schedule

Items	Jan, 2016	Feb	Mar	Apr	May	Jun	Jul	Aug	Sep
1. Public literature search on Fukushima debris status									
2. Public literature search on debris treatment methods									
3. Evaluate promising pyroprocess methods for the debris treatment									
4. Rough evaluation of the demonstration facility design and its cost/schedule									
5. Identify technical challenges									
6. Consideration on preferred metrics for nuclear waste transmutation									

## 2. Public literature search on Fukushima debris status

### Predicted debris location in 1F plants (based on SAMPSON code analysis)



Ref. 1: IRID Annual Research Report 2014 (in Japanese)

Ref. 2: M. Naito et. al., "Accident progress analysis by SAMPSON code", Shizuoka Univ., 2015  
Fall Meeting of the Atomic Energy Society of Japan, September 11, 2015 (in Japanese)

## 2. Public literature search on Fukushima debris status

### Assumed debris compositions for pyro-process treatment

Region		Core region				Lower plenum region		Pedestal / drywell region						
compositions		UO <sub>2</sub>	(U,Zr)O <sub>2</sub>	SUS-Zry alloy	Zr/Fe boride	SUS	(U,Zr)O <sub>2</sub>	(U,Zr)O <sub>2</sub>	(Zr,U)SiO <sub>4</sub>	CaAl <sub>2</sub> Si <sub>2</sub> O <sub>8</sub>	CaSiO	SiO	Fe-Si	Fe-Cr-Ni alloy
Fuel amount (t-UO <sub>2</sub> )	1F-1	0				0		79						
	1F-2	54						54						
	1F-3	0				11		99						
Predicted amounts		Major components			Minor components	Major components		Major components			Minor components			
Assumed compositions for pyro-process treatment		<p style="text-align: center;"><b><u>Core Debris</u></b></p> <p style="text-align: center;">(U,Zr)O<sub>2</sub> + SUS-Zry alloy</p>						<p style="text-align: center;"><b><u>MCCI Debris</u></b></p> <p style="text-align: center;">(U,Zr)O<sub>2</sub> + (Zr,U)SiO<sub>4</sub> + CaAl<sub>2</sub>Si<sub>2</sub>O<sub>8</sub> + CaSiO + Fe-Si + Fe-Cr-Ni alloy</p>						

MCCI: Molten Core Concrete Interaction

### 3. Public literature search on debris treatment methods

#### Comparison of major debris treatment methods (1/2)

Category		Pyro (metal)	Pyro (metal)	Pyro (oxide)
Base process		CaCl <sub>2</sub> electro-reduction (ER) + LiCl-KCl electro-refining (ER)	LiCl electro-reduction (ER) + LiCl-KCl electro-refining (ER)	Cl <sub>2</sub> gas chlorination + NaCl-CsCl electro-winning (EW)
Potential for debris treatment		+ Could reduce + Higher reduction potential + Some experiments on debris  -- less experience than LiCl electro-refining	+ Could reduce + Some experiments on debris  -- Li <sub>x</sub> Zr <sub>y</sub> O <sub>z</sub> produced as an impediment to electro-refining	+ Could dissolve debris + Some experiments on debris
Criteria in this study	Debris dissolubility	Yes	Yes (except Li <sub>x</sub> Zr <sub>y</sub> O <sub>z</sub> )	Yes
	Pu recovery with U and MA	Yes	Yes	No
	Compatibility to metal fuel recycling	Yes	Yes	No
	Secondary waste amount	Large	Small	Large
Assumed process in this study		✓ [Option 1]	✓ [Option 2]	--

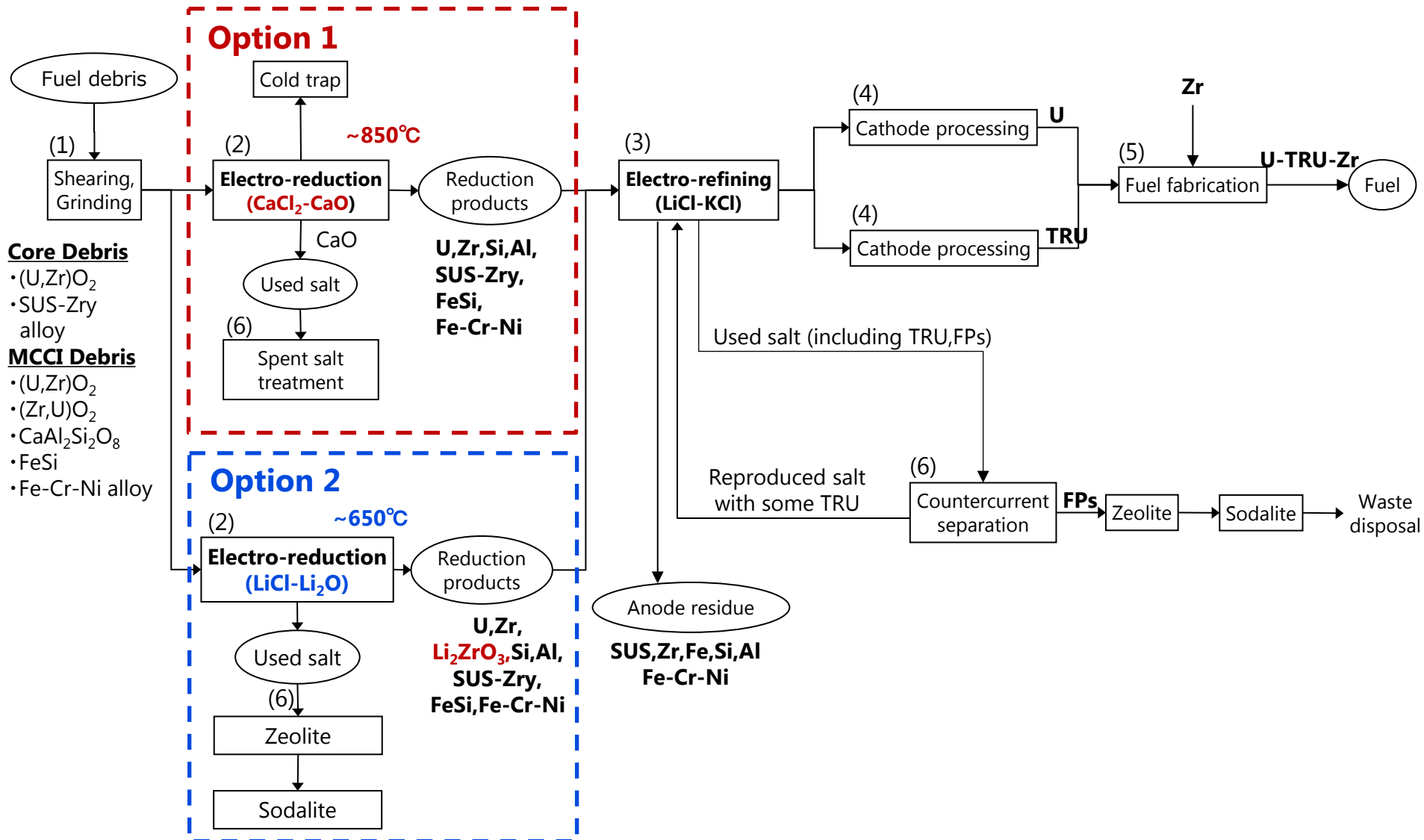
# 3. Public literature search on debris treatment methods

## Comparison of major debris treatment methods (2/2)

Category		Pyro (oxide)	FLUOREX	PUREX
<b>Base process</b>		molybdate melt + pyro-, or aqueous-process, etc	F <sub>2</sub> gas volatilization + solvent extraction	Nitric acid solution + solvent extraction
<b>Potential for debris treatment</b>		+ Could dissolve debris + Some experiments on debris  -- Unknown for MCCI Debris	+ Could dissolve debris + Some experiments on debris  -- Unknown for MCCI Debris	-- Difficult to dissolve debris + Some experiments on debris
<b>Criteria in this study</b>	<b>Debris dissolubility</b>	Core debris: maybe yes MCCI debris: unknown	maybe yes	No
	<b>Pu recovery with U and MA</b>	No	Yes	No
	<b>Compatibility to metal fuel recycling</b>	No	No	No
	<b>Secondary waste amount</b>	(unknown)	(unknown)	Larger
<b>Assumed process in this study</b>		--	--	--



# 4. Pyroprocess for debris treatment



## 4. Pyroprocess for debris treatment

### Basic thoughts on debris treatment process

- The most important thing for debris treatment process is high dissolution capability of various and complicated debris compounds such as MCCI debris.
- There are many unknown factors in every technologies, so far.
- Among them, pyroprocess seems to be one of the promising technologies for debris treatment, and it can be directly used also for metal fuel fast reactor cycle in the future as well.
- $\text{CaCl}_2$  electro-reduction vs  $\text{LiCl}$  electro-reduction for debris treatment
  - ✓ Reduction capability :  $\text{CaCl}_2 > \text{LiCl}$
  - ✓ Past experiences (on fuel cycle research) :  $\text{CaCl}_2 < \text{LiCl}$
  - ✓ Major technical concerns (refer to the appendix for details)
    - $\text{CaCl}_2$ : low reduction speed at the inner area of the debris, higher operating temperature, increased used salt waste
    - $\text{LiCl}$  : production of  $\text{Li}_2\text{ZrO}_3$  that impedes the process
- In this study, “ $\text{CaCl}_2$  electro-reduction +  $\text{LiCl}$ - $\text{KCl}$  electro-refining (Option 1)” is selected as the first option, so far, from a viewpoint of higher reduction capability.

# 5. Technical challenges

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## (1) Shearing and grinding

- Clarify shearing/grinding conditions based on the debris characteristics
- Confirm durability of the edge against high hardness borides
- Evaluate applicability of the shredder system that had been developed for oxide spent fuel
- Establish the pretreatment method of the debris for the electro-reduction process
- Confirm the loss rate, especially for fuel materials in the process

# 5. Technical challenges

## (2) Electro-reduction

### CaCl<sub>2</sub>

- Establish the electro-reduction condition
  - Debris material form (powder, sintering, ingot, etc)
  - Mitigate low reduction speed at the inner area of the debris
- Establish the countermeasures against high temperature operation (850°C)
  - Select proper structural materials
  - Reduce operating temperature using CaCl<sub>2</sub>-based low-melting salts, etc
- Clarify the treatment of the residual salt (CaCl<sub>2</sub>) in the reduced product
- Confirm material balance in the process

### LiCl

- Establish the electro-reduction condition
  - Debris material form (powder, sintering, ingot, etc)
  - Mitigate production of lithium zirconate (Li<sub>2</sub>ZrO<sub>3</sub>)
- Develop separation technology of lithium zirconate (Li<sub>2</sub>ZrO<sub>3</sub>)
- Confirm material balance in the process

## 5. Technical challenges

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### (3) Electro-refining

- Clarify electro-winning behavior of the reduced product alloyed with debris constituents such as Fe, Si, Al, Zr etc.
- Establish the treatment of the uranium oxide produced by lithium zirconate in the case of LiCl electro-reduction
- Confirm material balance in the process

### (4) Cathode processing

- Establish the distillation condition of salt where some debris constituents are mixed in.
  - Solid cathode (U),      - Liquid cathode (TRU)
- Confirm material balance in the process

### (5) Fuel fabrication

- Establish the injection casting conditions (temperature, time, atmosphere, pressure, etc.) for metal fuel with some debris constituents as impurities
- Establish the criteria on the allowable inclusion of debris constituents in the fuel product
- Confirm material balance in the process

# 5. Technical challenges

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## (6) Used salt treatment

- Develop the recycling technology for the used  $\text{CaCl}_2$  salt treatment
- Develop separation technology of the reduced debris constituents (Fe, Si, Al, Zr, etc.) from used salt if they are dissolved in.
- Clarify influence of the debris constituent inclusion in the conventional used-salt treatment processes and establish its countermeasures if needed.
- Confirm material balance in the process

## 6. Discussions

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- $\text{CaCl}_2$  electro-reduction (Option 1)
  - ✓ Can “ $\text{CaCl}_2$  electro-reduction” really reduce most of the debris components including MCCI?
  - ✓ Is there any reasonable method for used  $\text{CaCl}_2$  salt treatment to reduce the process wastes?
  - ✓ Does the combination of  $\text{CaCl}_2$  salt process (reduction) and  $\text{LiCl-KCl}$  salt process (refining) has technical issue?
  - ✓ Are there any other critical issues?
  
- $\text{LiCl}$  electro-reduction (Option 2)
  - ✓ How to solve the  $\text{Li}_2\text{ZrO}_3$  issue?
  - ✓ Are there any other critical issues on Core Debris and MCCI debris treatments?
  
- $\text{LiCl-KCl}$  electro-refining (common to Option 1 & 2)
  - ✓ How do you think about the behavior of the reduced product alloyed with debris constituents such as Fe, Si, Al, Zr, etc?
  
- Are there further improvement ways on pyroprocess for debris treatment?

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# Backup slides

# Technology roadmap (draft) on pyroprocess for debris treatment

Only for discussion

Years	1	2	3	4	5	6	7	8	9	10	11	12	13	14	15	16	17	18	19	20	21	22	23	24	25	26	27	28	29	30	
<b>Development Phase</b>	<b>Principle verification</b>				<b>Laboratory scale</b>			<b>Engineering actual proof</b>								<b>Demonstration facility for the debris treatment</b>															<b>Operation</b> ▾
(1) Shear pulverization					Several 100g scale																										
(2) Electro-reduction	Several 10g scale				Several 100g scale			<b>Process selection</b>																							
○LiCl								▽ Several kg scale																							
○CaCl <sub>2</sub>	Several 10g scale				Several 100g scale																										
(3) Spent salt treatment																															
(4) Electro-refining																															
(5) Cathode processing																															
(6) Fuel fabrication																															
																Basic design			Detail design			Approval of design and construction plan			Construction						