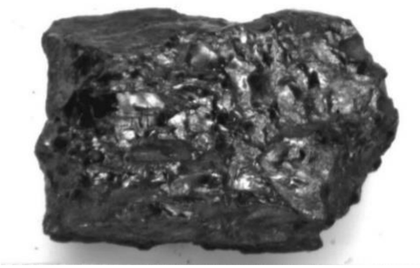
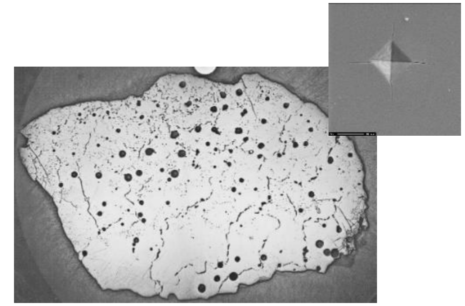




Fuel debris characterization for Decommisiioning of Fukushima Daiichi NPS

Tadahiro WASHIYA

July 3, 2017

Collaborative Laboratories for Advanced Decommissioning Science (CLADS),

Japan Atomic Energy Agency (JAEA)

International Research Institute for Nuclear Decommissioning (IRID)

This paper includes part of the results obtained under the research program entrusted to International Research Institute for Nuclear Decommissioning (IRID) including Japan Atomic Energy Agency (JAEA) by the Agency for Natural Resources and Energy, Ministry of Economy, Trade and Industry (METI) of Japan.

Introduction

- **JAEA has been studied characterization of fuel debris and MCCI product for Fukushima Daiichi NPP (1F).**
  - Obtained fundamental data by simulated fuel debris and TMI-2 sample with considering 1F distinctive reactions
  - Estimated 1F debris properties as mechanical and chemical
  - Created the " Estimated properties list " of the 1F debris

⇒ Contribute to preparation and planning for 1F decommissioning.

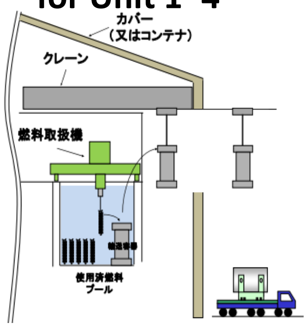
- **1F debris sampling will be performed in the near future, and JAEA is conducting preparation of the analysis.**
  - To consider purpose of sample analysis
  - To confirm debris properties by 1F sample analysis
  - To confirm damaged condition in 1F reactor core by the results
  - International cooperation will be effective

⇒ The 1F' debris sample will more specifically contribute to the defueling, storage, conditioning and disposal, and also to study on SA research and accident management.

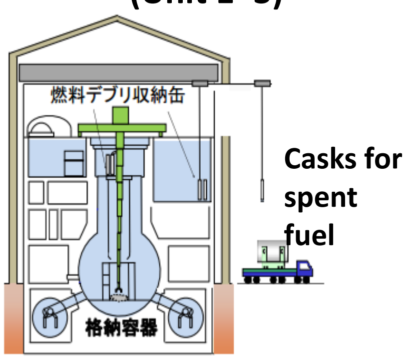
1F Decommissioning Schedule

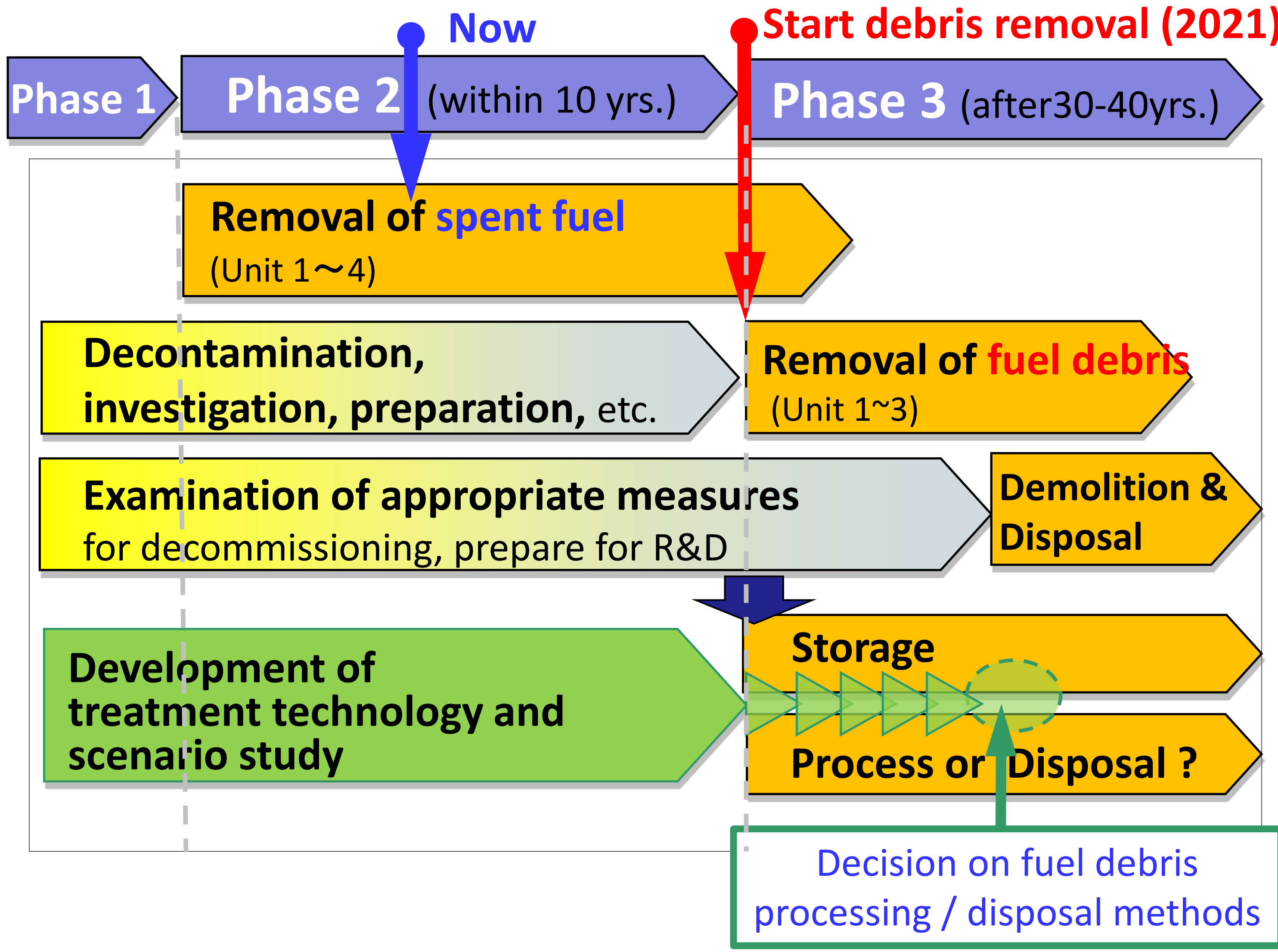
Mid-and-Long Term Roadmap

Removal of spent fuel for Unit 1~4



Removal of fuel debris (Unit 1~3)



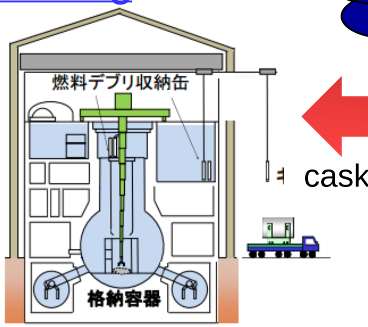


Purpose of debris characterization

Decommissioning

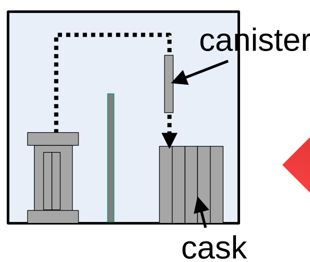
Defueling

ex. Submerged defueling



Storage

ex. Wet storage



Treatment

ex. U/Pu recovery and vitrification

Disposal

Decision of Tools & Methods

Consideration of Pre-treatment

Consideration of Treatment

Debris' property

Mechanical property
Hardness, Fracture toughness, etc.

Thermal property
Thermal conductivity, Melting temperature, etc.

Physical property
Density, Drying characteristic, etc.

Chemical property
Acid-solubility, antioxidant property, etc.

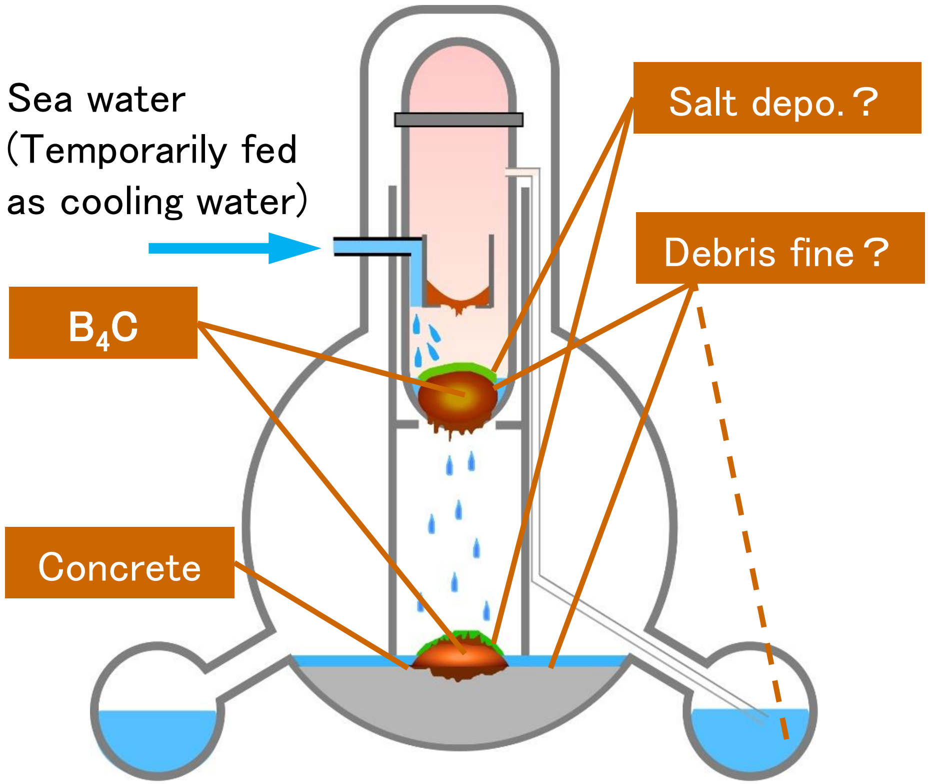
Specific effects on 1F debris property

The 1F debris, 1F specifically conditions are considered as following.

- Effect of sea water (alternative coolant) ～salt deposition on the debris surface？
- Effect of B_4C as control rod material ～ boride' chemical form and distribution in the debris
- Effect of concrete interaction (MCCI)
- Other effects: Pu, Gd（BP）, leaching,
  FP chemical form and etc.

The property is estimated by simulated debris（Solid phase, Components, Mixing rate, Mechanical and thermal proierties)

Contribute to debris defueling (tool design, criticality safety control, storage and management)



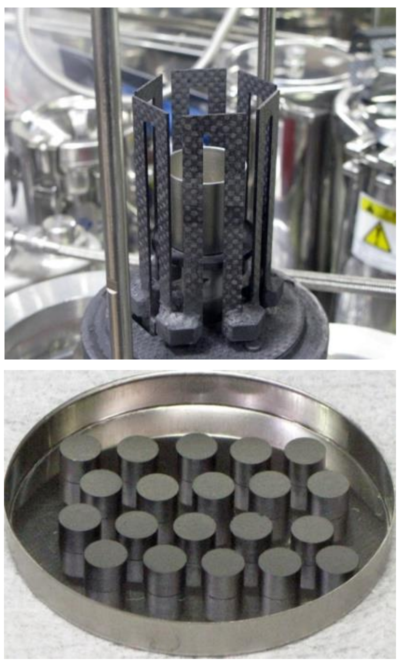
Typical perspective view of 1F reactor

Fabrication of sim. fuel debris

(1) Sintering

$yUO_2 + (1-y)ZrO_2 \rightarrow (U_yZr_{1-y})O_2$

- Same as fuel pellet fabrication
- Mixture, Crush, Pelletize, Sintering（Ar, ~1730℃）
- Additive: Gd_2O_3, Fe_2O_3, CaO
- With control oxygen potential



(2) Arc dissolution

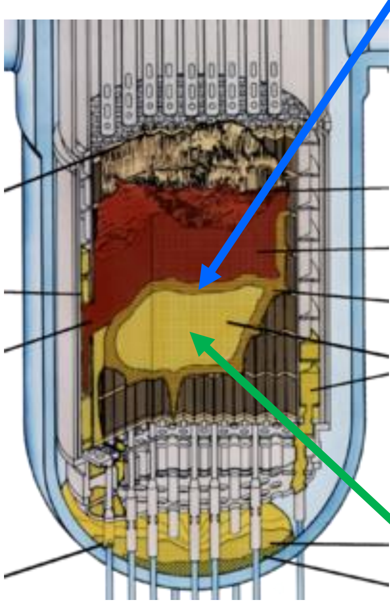
- Entire uniform heating
- Required metal rich conditions
- Simulated Core Debris , MCCI product
- ～5 g / sample

(3) Heating by light collection

- Concentrating light as heat soce
- Erosion under thermal gradient conditions
- Various atmosphere gas
- Small scale MCCI: Melting of $(U,Zr)O_2$

Effect of cooling rate on $(U,Zr,Fe)O_2$

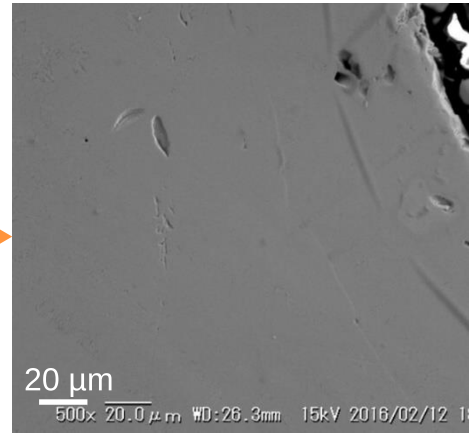
Upper crust

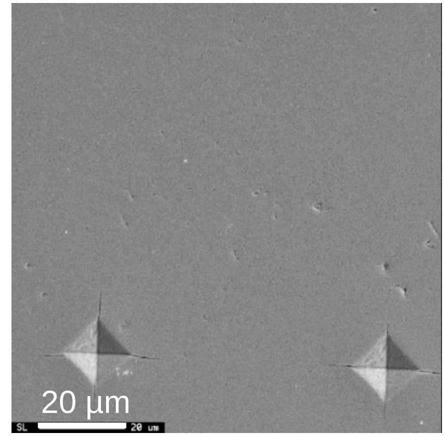


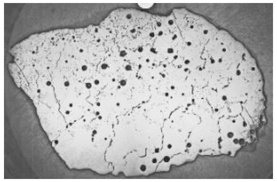
Melting pool

Sim. debris | **TMI-2 debris**

Rapid cooling from molten state



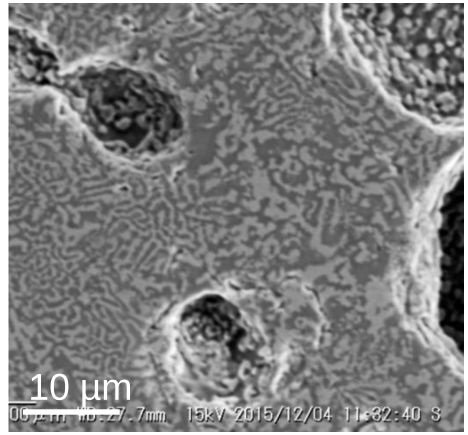


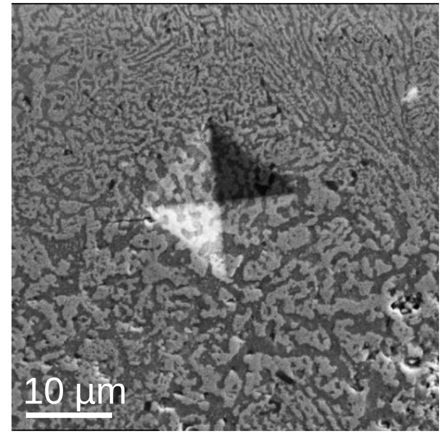


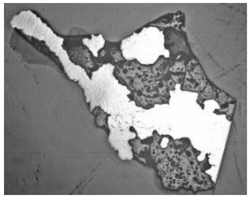
Upper crust (Rapid cooling)

Cubicle single phase

Sintering at 1600℃





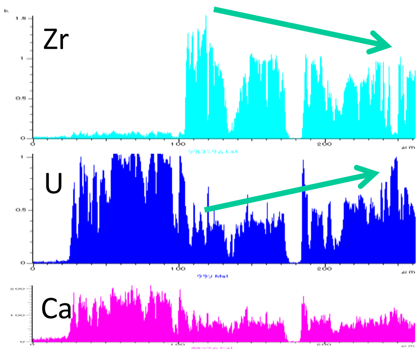


Melting pool (Annealed)

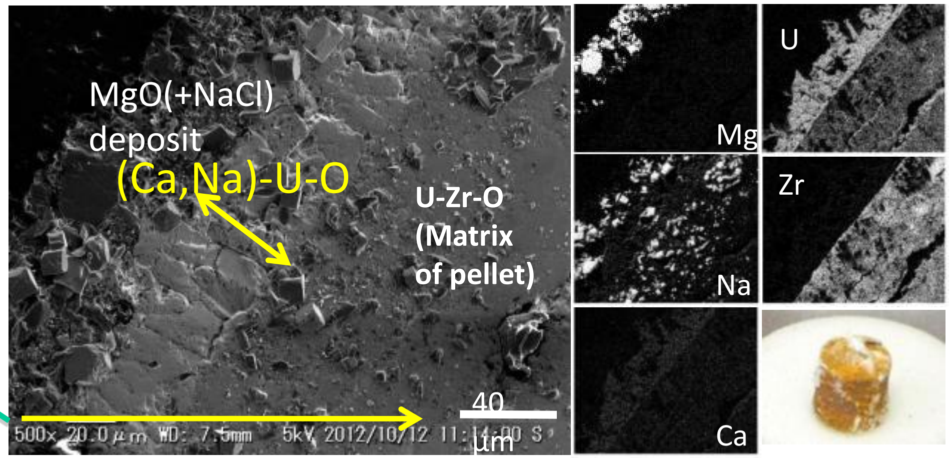
Mixture of U-rich Cubicle and Zr-rich Tetragonal

Sea salt and B_4C effect on simulated debris

(1) $(U_{0.4},Zr_{0.6})O_2$ – Sea water reaction in high temp.



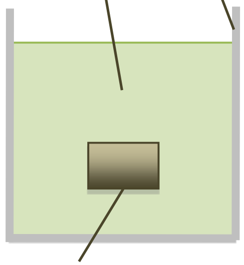
Elemental profile



SEM images on the (U0.4Zr0.6) O_2 pellet Heat-treated with salt at 1275K for 12h under air flow.

Aluminum crucible

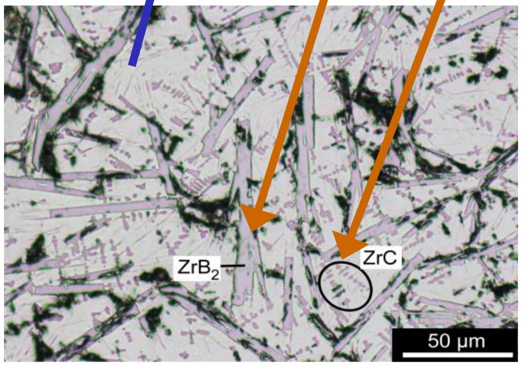
Salt filling



Simulated debris pellet

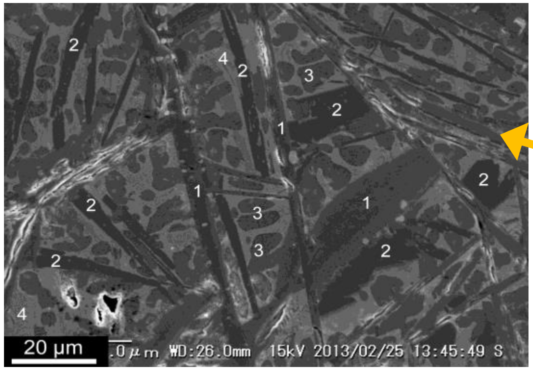
(1) B_4C reaction in high temp.

(Fe,Cr,Ni)$_2$Zr ZrB$_2$ ZrC

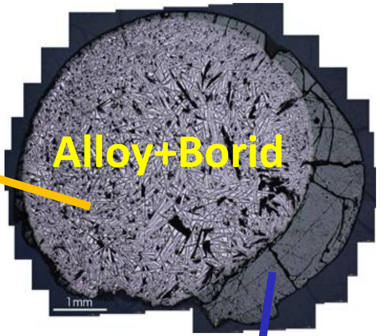


B_4C/SS/Zr Mixture

1: ZrB_2
2: (Fe,Cr,Ni)$_2$B
3: Fe-Cr-Ni
4: Fe-Cr-Ni-(Zr,U)



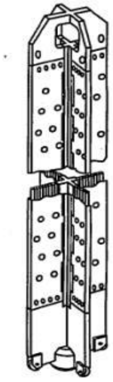
B_4C/SUS/Zr/(U,Zr)O_2 Mixture



(U,Zr)O_2

Mixture of B_4C/SS/Zr/(U,Zr)O_2

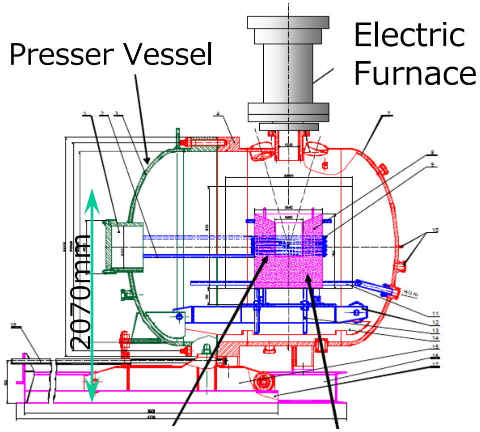
Arc melting

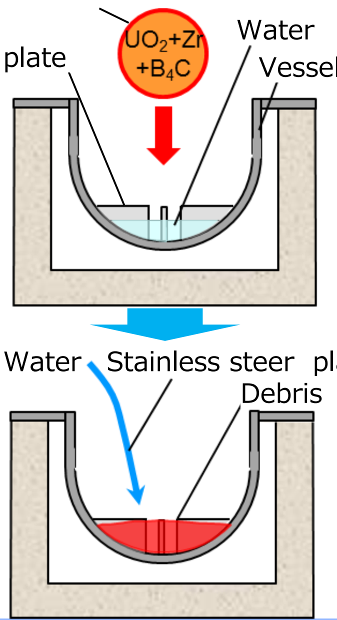


Control rod

Molten corium and metal reaction

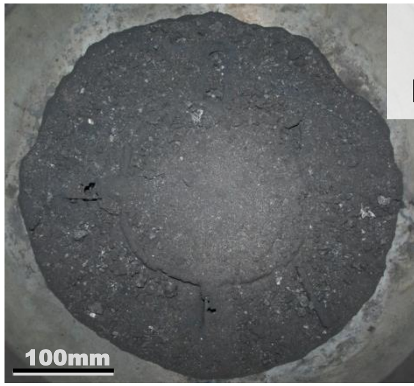
Collaboration with Toshiba and National Nuclear Center of the Republic of Kazakhstan (NNC)



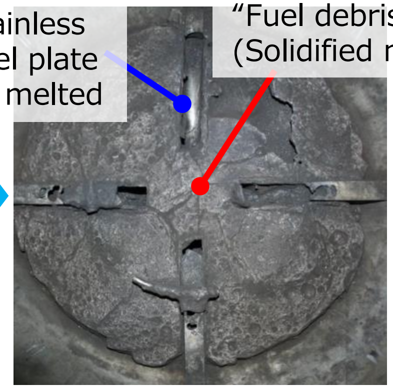


Slow-cooling conditions

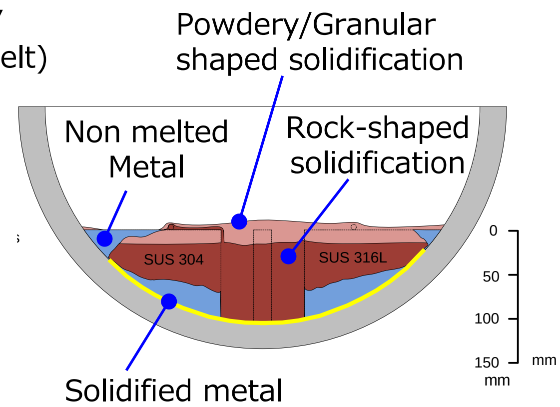
After test



After powdery debris removed

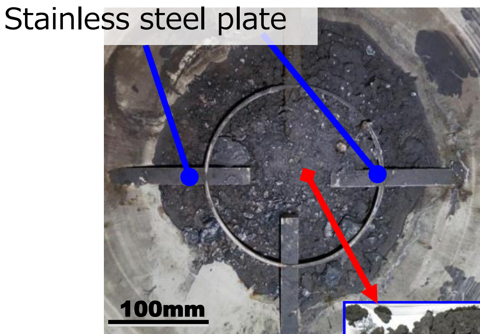


Cross section



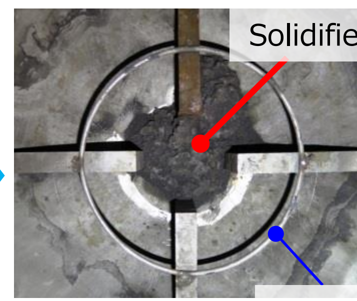
Water-cooling conditions

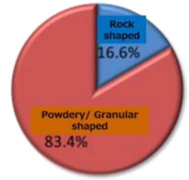
After test



"Fuel debris" (Powdery/Granular shaped solidification)

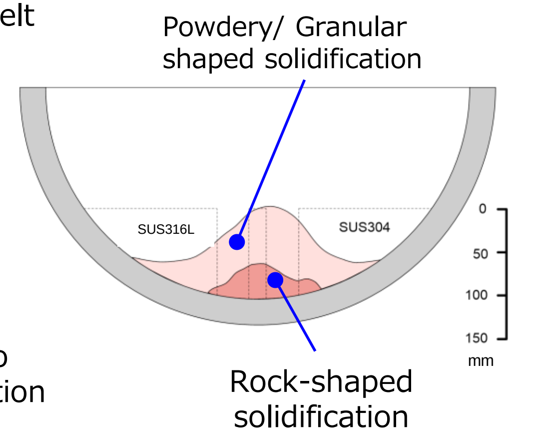
After powdery debris has removed





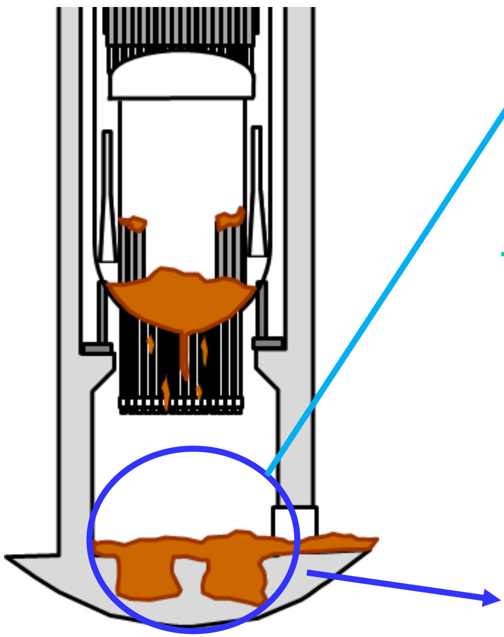
Weight ratio of solidification

Cross section

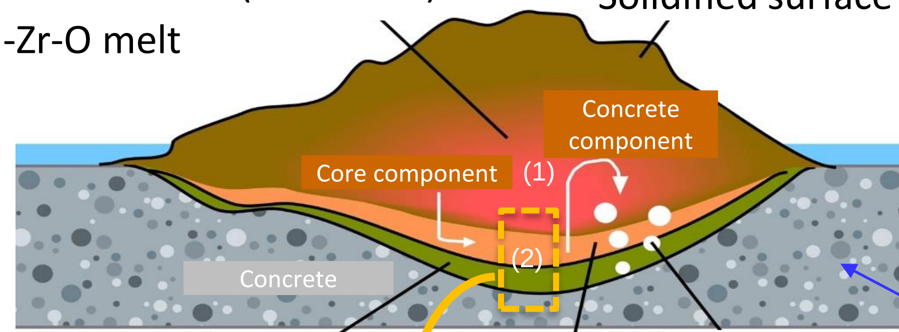


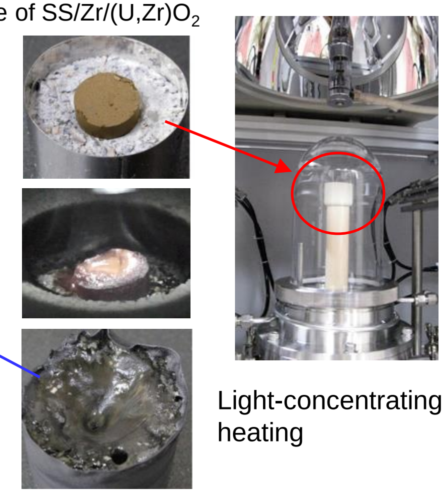
MCCI effects on pedestal concrete

Image of the sediment of MCCI product

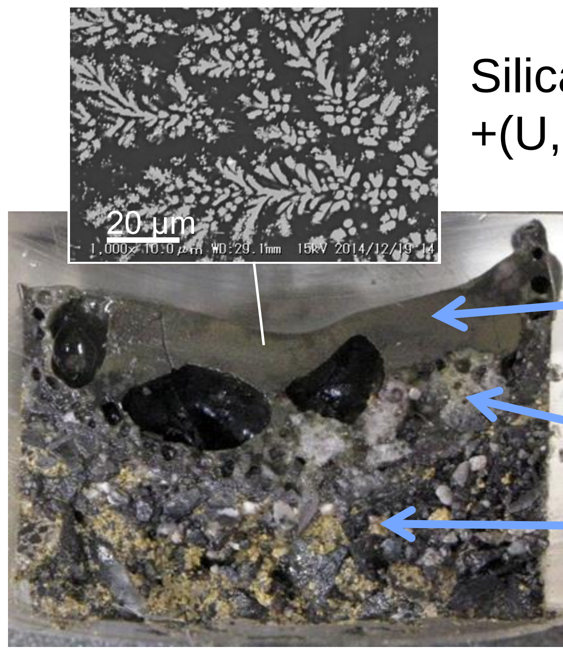


Layered-structure image

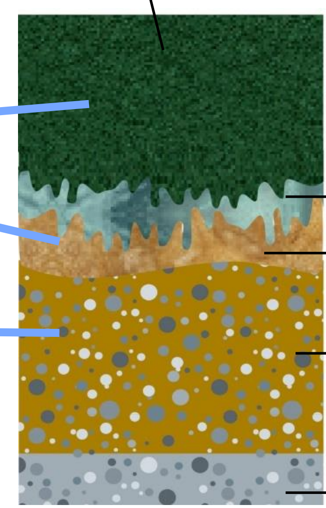




Light-concentrating heating



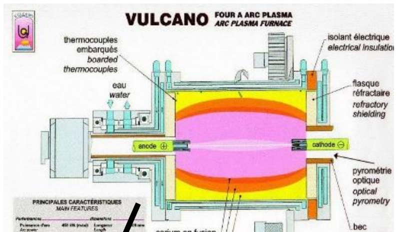
Silicate glass (containing 3-4 wt%-U) +(U, Zr, Ca) O_2 precipitation

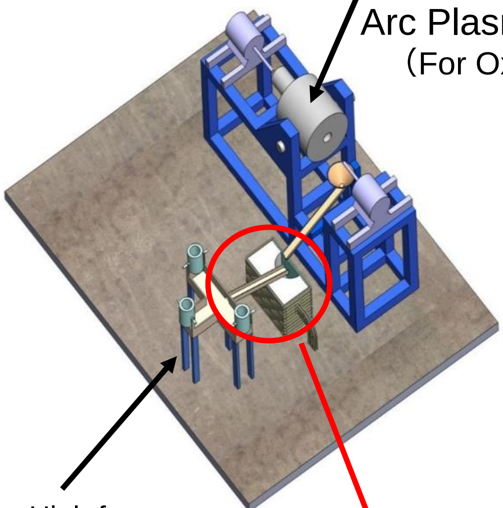


Cross section after melting reaction
(Concrete piece with a diameter of 25 mm)

MCCI product in engineering scale test

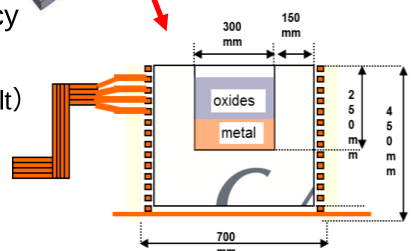
Collaboration with JAEA and French Atomic Energy and Alternative Energies Commission (CEA)



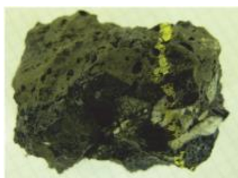


Arc Plasma furnace
（For Oxide melt）

High frequency furnace
（for Metallic melt）



Engineering scale MCCI Test Facility : VULCANO @CEA



MCCI sample

Calculation results by TOLBIAC-ICB

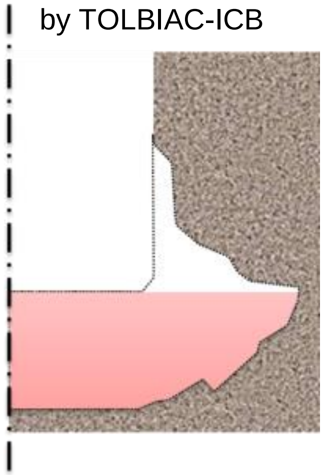
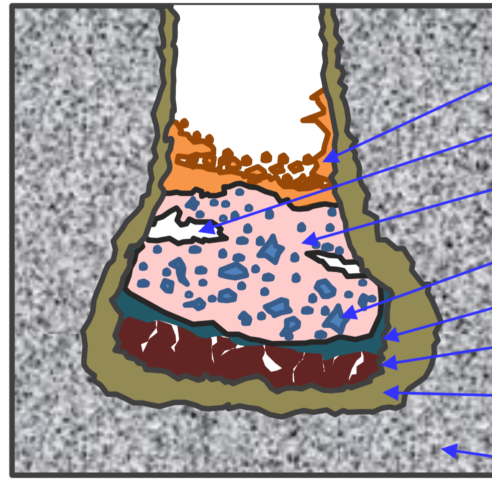
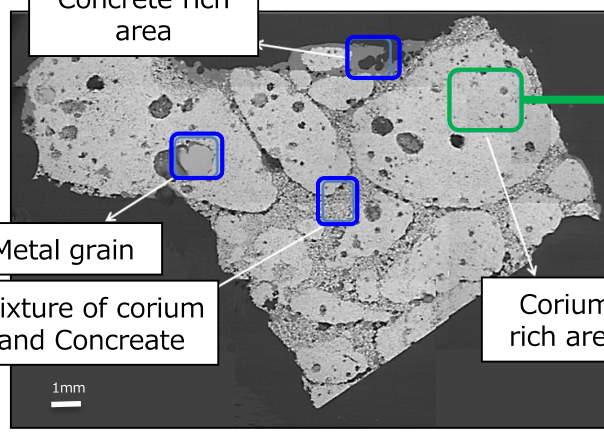


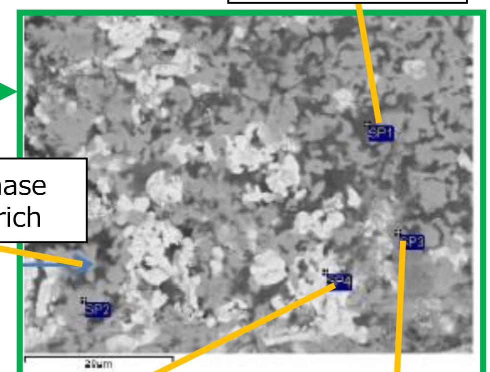
Image of cross section of MCCI



Upper crust
Cavity
Corium oxide
Metallic extraction
Metallic Layer
Lower Crust
Thermally-degraded concrete
Intact Concreate



Concrete rich area
Metal grain
Mixture of corium and Concreate
Corium rich area



Oxide phase with Si rich
Oxide phase with Zr rich
Oxide phase with U rich
Metallic phase with Fe rich

SEM/EDS analysis

Chemical alternation of LFCM/Che-NPP

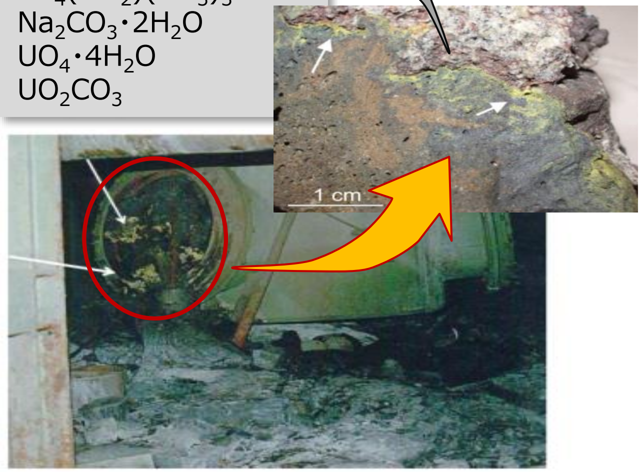
Collaboration with JAEA and Institute for Safety Problems of Nuclear Power Plants of NASU (ISP-NPP)

Chernobyl's experience

◆ Chemical alteration of debris

- Chernobyl's "LFCM : Lava-like Fuel-Containing Materials" is still leaving in the reactor area, hence LFCM slowly react with moisture and air. 1)
- Secondary chemical species are generated on the surface, including dissolvable uranium.

$Na_3H(CO_3)_2 \cdot 2H_2O$
$UO_3 \cdot 2H_2O$
$Na_4(UO_2)(CO_3)_3$
$Na_2CO_3 \cdot 2H_2O$
$UO_4 \cdot 4H_2O$
UO_2CO_3

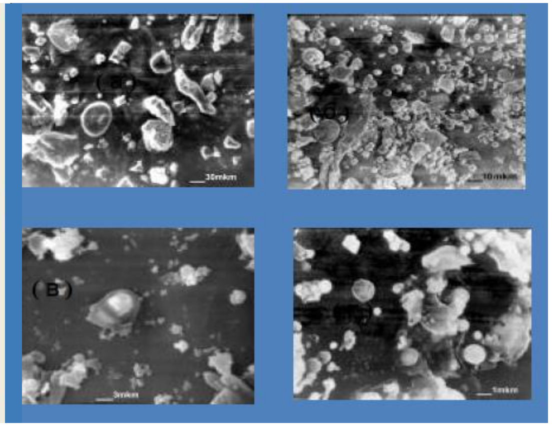


Uranyl- phases generated on the surface of Chernobyl's "Lava"

◆ Aerosol generation

- Aerosol is generated on the surface of LFCM continuously. 2)
- Major radio nuclides are $^{137}Cs/^{241}Am$, $^{241}Am/^{154}Eu$, $^{137}Cs/^{154}Eu$

<u>Concerning for Fukushima Daiichi NPP</u>
- Potential of similar alteration on the fuel debris & MCCI production in 1F conditions.
- Treatment of the aerosol generated by the fuel debris & MCCI production.



Aerosol generated from LFCM

1) B. Burakov "Material Study of Chernobyl "Lava"and"Hot"Particles", International Experts'Meeting on Decommissioning and Remediation after a Nuclear Accident ,Vienna,Austria,28 January-1 February 2013.

2) http://www.jsme.or.jp/pes/Research/A-TS08-08/index02.html

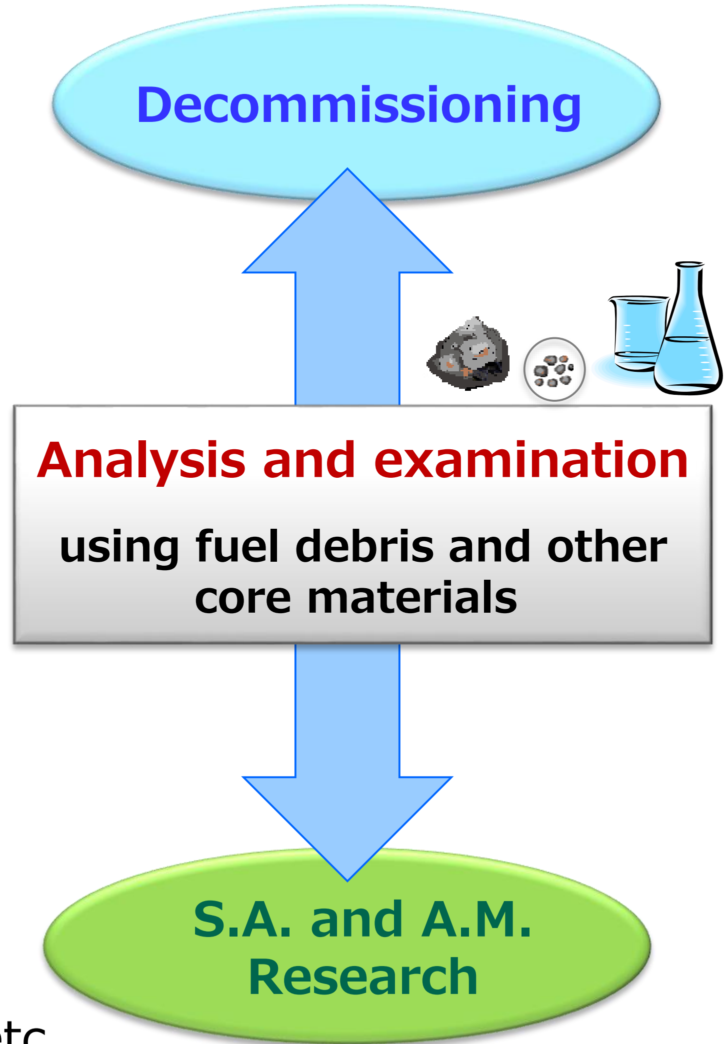
Purpose of debris analysis and examination

◆ Contribution to decommissioning

- **To understanding the damaged state inside the reactors;** quantity, shape, composition, etc.
- **To understanding the contamination;** radiation, nuclides, chemical form, etc.
- **To understanding the safety;** criticality, hydrogen, corrosion, etc.
- **To examining the defueling;** criticality, tools, method, etc.
- **To storing and storage**
- **To reatment and disposal**

◆ Contribution to SA and AM

- **To investigating the accident progression;** in/ex-vessel phenomena, containment failure, fission product behavior, and source term, etc.
- **To investigating the system reliability;** coolability , containment, corrosion and erosion, etc.



International Joint Research Project "Preparatory Study on Analysis of Fuel Debris (PreADES)" will be launched by OECD/NEA/CNSI

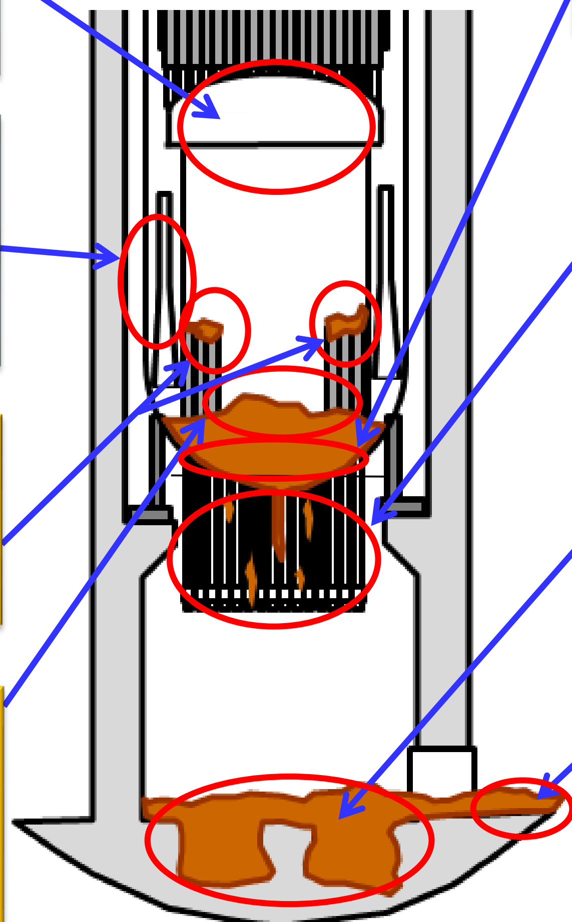
Image of 1F core conditions and analysis purpose

Region A : Upper Structure
(Purpose)
Surface contamination survey, Radiation estimation, Study of decontamination method

Region B : Shroud
(Purpose)
Surface contamination survey, Radiation estimation, Study of decontamination method

Region C : Core part residue
(Purpose)
Evaluation of Residual volume, Residual form, Homogeneity and safety

Region D : Control Rod Guide Tube(CRGT)
(Purpose)
Evaluation of Residual volume, Residual form, Homogeneity and safety



Region E : RPV Lower Head
(Purpose)
Evaluation of Residual volume, Residual form, Homogeneity and safety

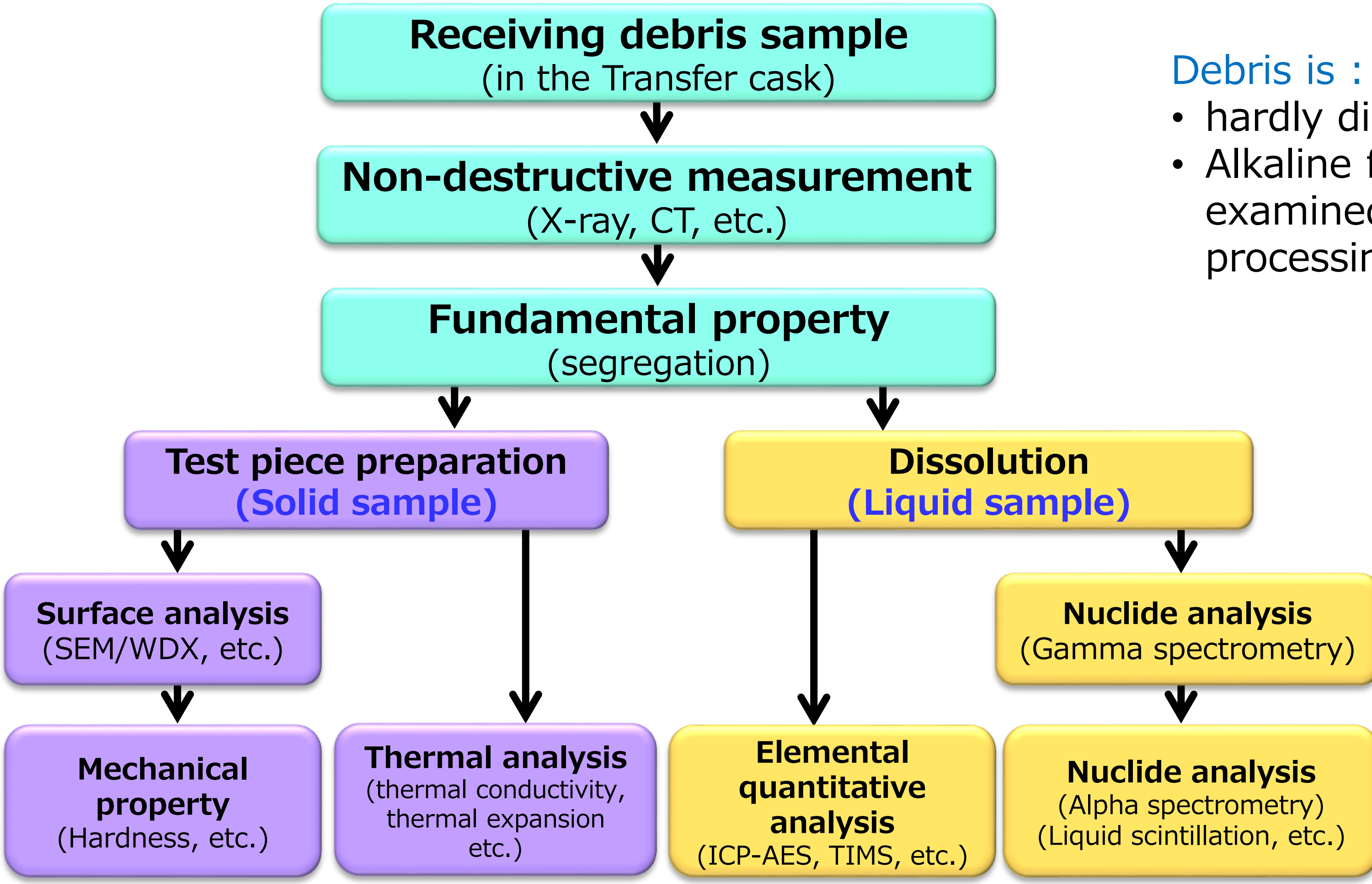
Region F : Control Rod
(Purpose)
Evaluation of Residual volume, Residual form, Homogeneity and safety

Region G : Pedestal Floor
(Purpose)
Evaluation of Residual volume, Residual form, Homogeneity and safety

Region H : D/W Floor
(Purpose)
Evaluation of Residual volume, Residual form, Homogeneity and safety

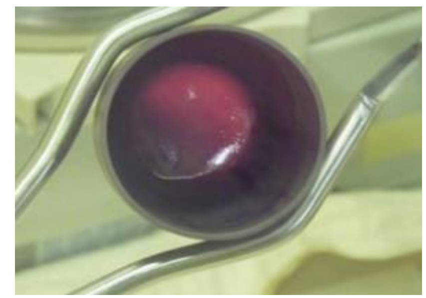
Analytical technology for fuel debris

- The analysis workflow considered to enable the efficient debris analysis.
- In this workflow, values of sample with unknown components are roughly analyzed by the non-destructive method and then the overall analyses is created.



Overall debris analysis workflow plan

Debris is :
- hardly dissoluble material
- Alkaline fusion method is being examined as a pre-analysis processing method.

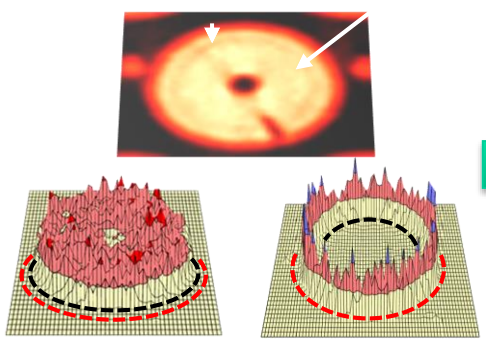


Debris dissolution
(by Alkaline fusion method)

The X-ray CT technology development

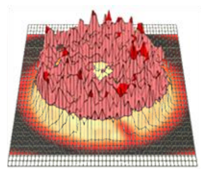
The X-ray CT test apparatus has been developed as a non-destructive examination device for examining the internal condition of subassembly (Budle Duct Interaction, etc) and for reflecting to soudness confirmation and fuel-life estimation, and installed in the extension facility of Fuel Monitoring Facility (FMF)

Density analysis：
Fuel clad tube（Red）、Fuel（Yellow）

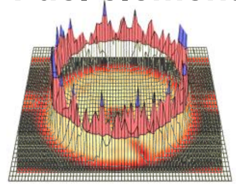


FPs from SF

Radioactive nuclide from structure



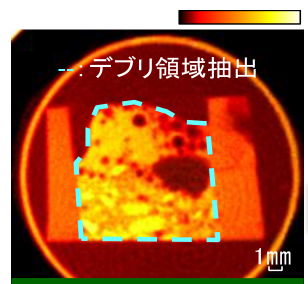
Fuel element



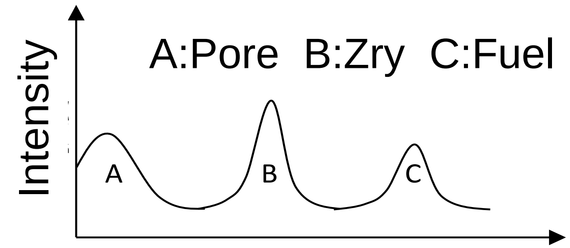
Structural materials

Cross sectional of gamma tomography

Density: low ⇔ high



X-ray CT of Simulated debri



Density histogram in the fuel debris area

- ◆ Construction of quantitative evaluation method of porosity due to pore, etc.
- ◆ Construction of fuel debris component identification method

Conclusion

■ Characterization on fuel debris

- **For 1F decommission, estimated debris properties is essential**
  - ➢ Fundamental data has been measured using simulated debris , etc..
  - ➢ Density, Mechanical properties (Hardness, Elastic modulus, Fracture toughness), and thermophysical properties (Thermal conductivity, Specific heat, Melting point).
- **1F's specifically conditions are investigated**
  - ➢ Seawater, B_4C as control rod material, Pu and concreate interaction was examined experimentally.
  - ➢ Chemical alteration on the debris at various conditions needs to be investigating.

■ Preparation of 1F debris analysis

- **Debris analysis will contribute to 1F decommissioning and SA research etc.**
  - ➢ Sample analysis help for understanding the damaged reactors core, for implementing the decommissioning work, for investigating the accident progression, and for safety analysis.
- **Debris analysis needs to be discussed internationally**
  - ➢ The 1F debris analysis can be used as expanding the knowledge and expertise on the severe accident internationally.

Thank you for your attention