The International Forum on the
Decommissioning of the Fukushima Daiichi NPS
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# JAEA's Efforts on Radioactive Waste/Fuel Debris Analysis

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(JAEA/CLADS)

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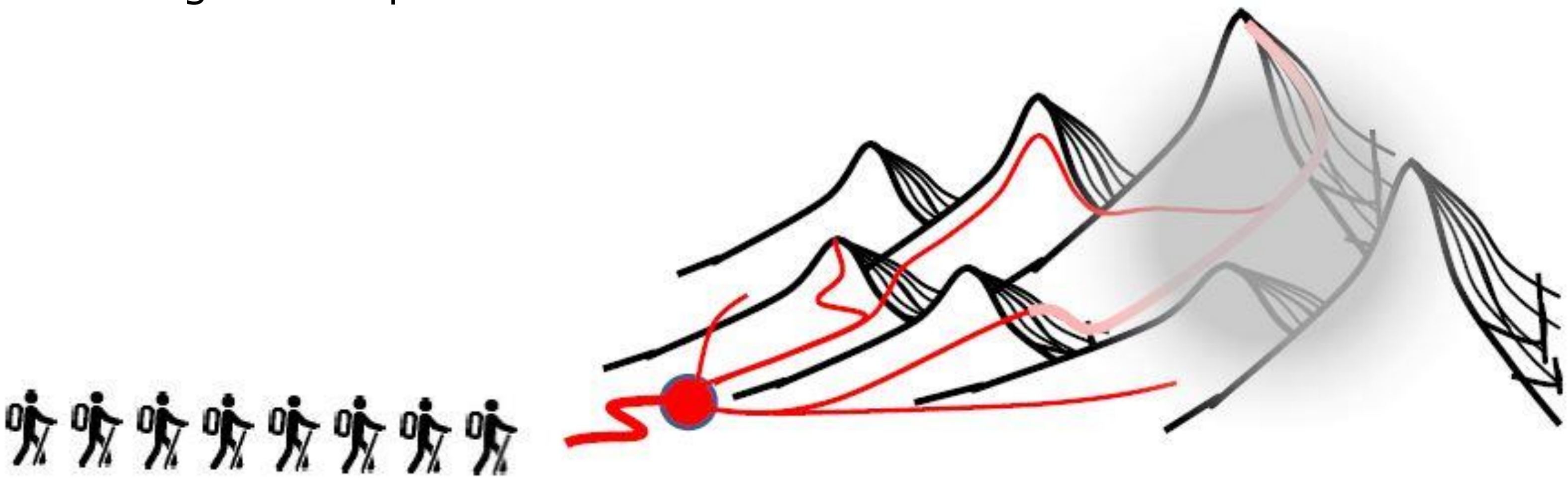
# Contents

# I. JAEA's mission for radioactive waste/fuel debris analysis

JAEA is a member of a mountaineering expedition aiming for unclimbed peaks. The best route has not been found yet, and JAEA will tackle together with our knowledge and experience.



JAEA takes on the role of Sherpa as an expert by research and practice related to analysis. JAEA will support the decommissioning work of TEPCO HD through knowledge and technology that can be used for safety and risk assessment.

JAEA's contributions
- Analysis technology, research and development, expert.
- Experience and achievements in analyzing radioactive waste, irradiated fuel, and TMI-2 debris
- Results of 1F sample analysis (waste, sediment, etc.)
- Design, construction and operation of the Okuma Analysis and Research Center

# I. JAEA's hot laboratories and human resource development

- As a human resource development base, training of young engineers and training and training of external engineers are being carried out. Established a collaborative laboratory with universities, implemented a cross-appointment system, and provided training for students


The Laboratory-1 for the radioactive material analysis and research facility has been completed, and the Laboratory-2 is under construction.

# I. Radioactive material analysis and research facility

- Under construction of Radioactive Material Analysis and Research Facility to understand the properties of radioactive waste and fuel debris on the adjacent to 1F。
- Radioactive materials with surface dose rate up to 1 Sv/h such as rubble, incinerated ash, and secondary waste from water treatment will be analyzed. And also ALPS-treated water will be analyzed by JAEA as third-party.
- High-level radioactive materials such as fuel debris will be analyzed.



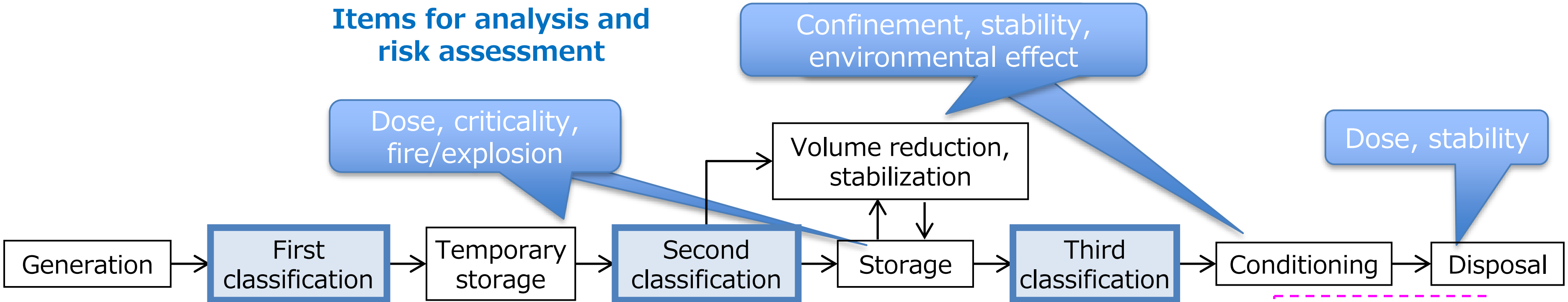
① Administrative building【 Operation started in March 2018 】

② **Laboratory-1※【2022/6/24 Completion. Scheduled to start analysis in 2022 】**

③ Laboratory-2※【Licensing /preparation for construction】

※ TEPCO applied for an implementation plan as part of the specified nuclear facility and supervised security. JAEA is in charge of design, construction and operation (analytical practice and facility operation).

# II. Analysis of radioactive waste Information and analysis required




Example on P.7, 8

## Needs for analysis

**Characterizing for storage**

- Surface dose rate
- Fissile material
- Water, radioactivity content

**Characterization for processing (stabilization)**

→ Dependent on waste and processing

Example: ALPS slurry

- Particle size distribution: for filtering
- Viscosity: for drying
- Radioactivity concentration: for device design
- Chemical composition

Characterization for decomposition

- Environmental chemical

**Characterization for processing (conditioning)**

→ Dependent on waste and processing

Characterization for disposal (including conditioning)

- Material
- Amount
- Radioactivity concentration
- Environmental chemical affecting radioactivity behavior

Feedback for early establishing management technologies

Data, result of evaluation

✓ Currently, radioactivity concentration is analyzed for first through third classification.

✓ Waste of Fukushima Daiichi features...

- Difficult to predict radioactivity composition including DTMs
- Enormous amount
- High dose rate

Example on P. 9

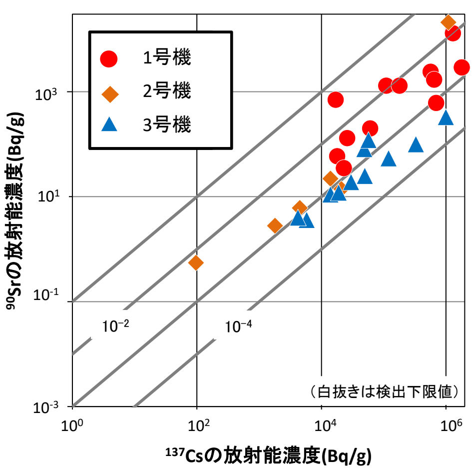
- Inventory estimation method
- Analysis planning method
- Analysis method for DTMs

Example on P. 10

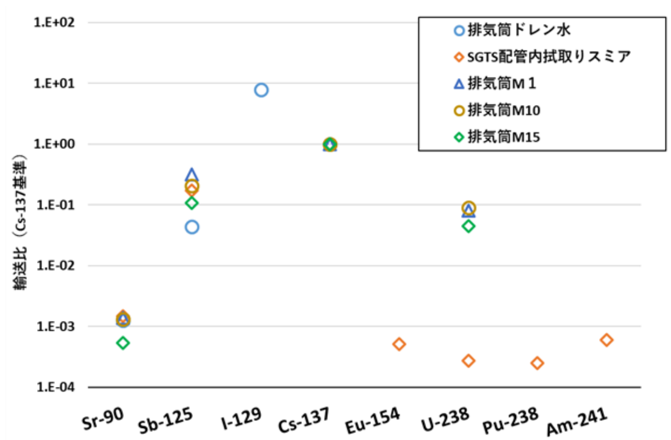
Feedback from data

# II. Analysis of radioactive waste   Results of analysis

- Since JFY2011, radioactive waste and contaminated materials have been analyzed, the number samples reached to 500.
- Rubble is one of major waste due to its large volume. Secondary waste from water decontamination including sludge/slurry, adsorbent and concentrate should be reduced its risk. These are the preferential targets of analysis.
- Data obtained is utilized to discuss correlation of activity concentration (1), classification of waste according to nuclide composition (2).
- Data is collected in a database “FRAnDLi” to open to public for use (3).
  - ✓ https://frandli-db.jaea.go.jp/FRAnDLi/index.php?country=e




(1) Correlation between $^{137}$Cs and $^{90}$Sr for rubble collected inside reactor buildings.*1




(2) Transport ratio of nuclides to stuck of 1/2 units, SGTS piping, drain water.


(3) Database “FRAnDLi”.

*1 : https://www.meti.go.jp/earthquake/nuclear/decommissioning/committee/osensuitaisakuteam/2017/05/3-04-03.pdf

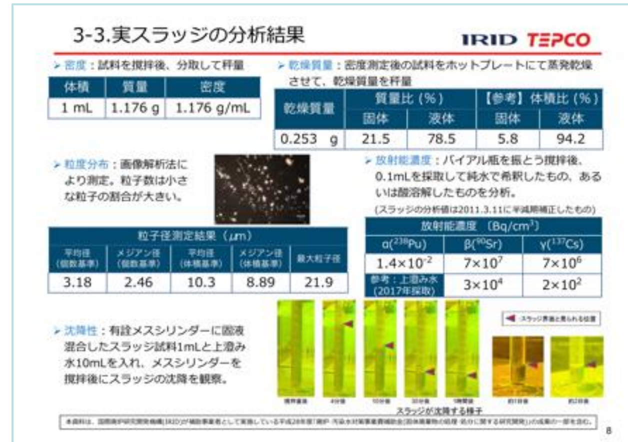
# II. Analysis of radioactive waste　Feedback to Fukushima Daiichi

- TEPCO utilizes analysis data including physical and chemical properties to reduce risk for waste storage by waste treatment as shown in report to government.

Sludge in contaminated water

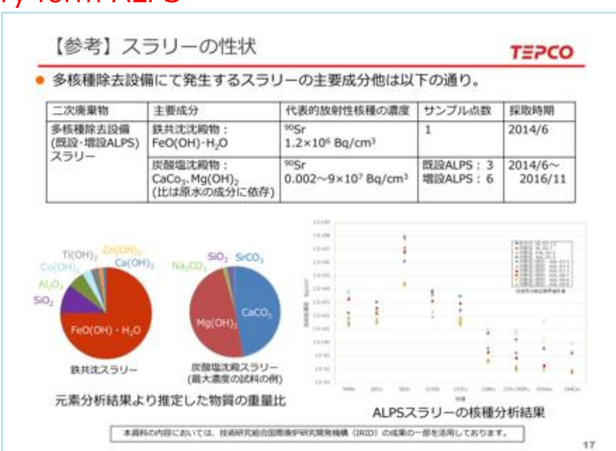

東京電力ホールディングス株式会社, “α核種除去に向けた検討状況について,” 廃炉・汚染水対策チーム会合／事務局会議（第86回）, 2021年1月28日.

Sludge from decontamination device




東京電力ホールディングス株式会社, “除染装置スラッジ、ALPSスラリーの安定化処理にむけた検討状況,” 特定原子力施設放射性廃棄物規制検討会 (第7回), 資料2, 2018年7月23日.

Slurry form ALPS



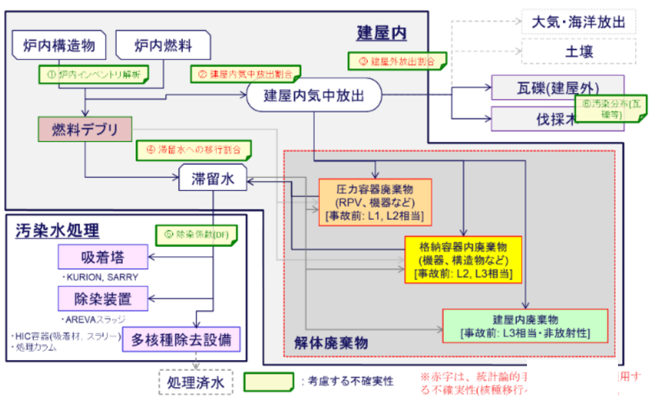

東京電力ホールディングス株式会社, “スラリー安定化処理に向けた設計について,” 特定原子力施設監視・評価検討会（第88回）, 資料1-2-1, 2021年2月22日.

Zeolite contained in sandbag

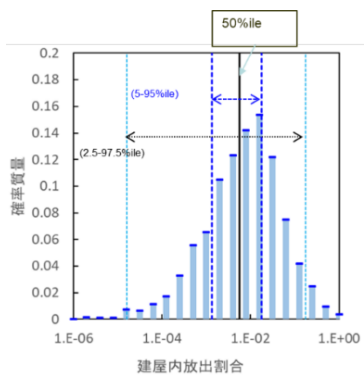

東京電力ホールディングス株式会社, “プロセス主建屋における地下階環境調査の結果について,” 廃炉・汚染水・処理水対策チーム会合／事務局会議（第93回）, 2021年8月26日.

# II. Analysis of radioactive waste Methods of inventory estimation and planning

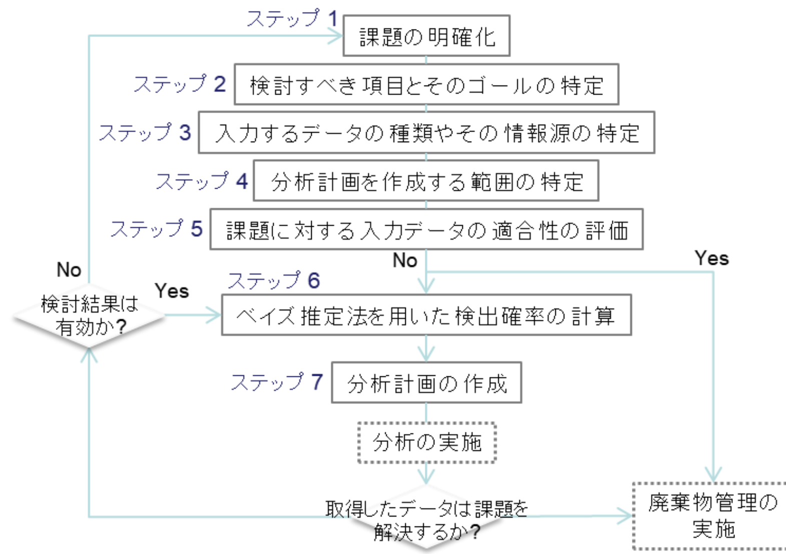
- To estimate inventory of various waste, estimation model is being developed. Model includes parameters of transport of radioactive nuclides, which is derived from analysis data. Analytical concentration is normalized with fuel composition and statistically treated to obtain normal distribution parameters, and uncertainty is evaluated with help of Bayesian approach. (1)。
- To make efficient analysis plan for waste of uncertain, DQO process is combined with Bayesian statistics to set number of samples for classifying various waste, which should be conformed to future conditioning/disposal (2).




Calculation model[*1]




Parameter[*2]



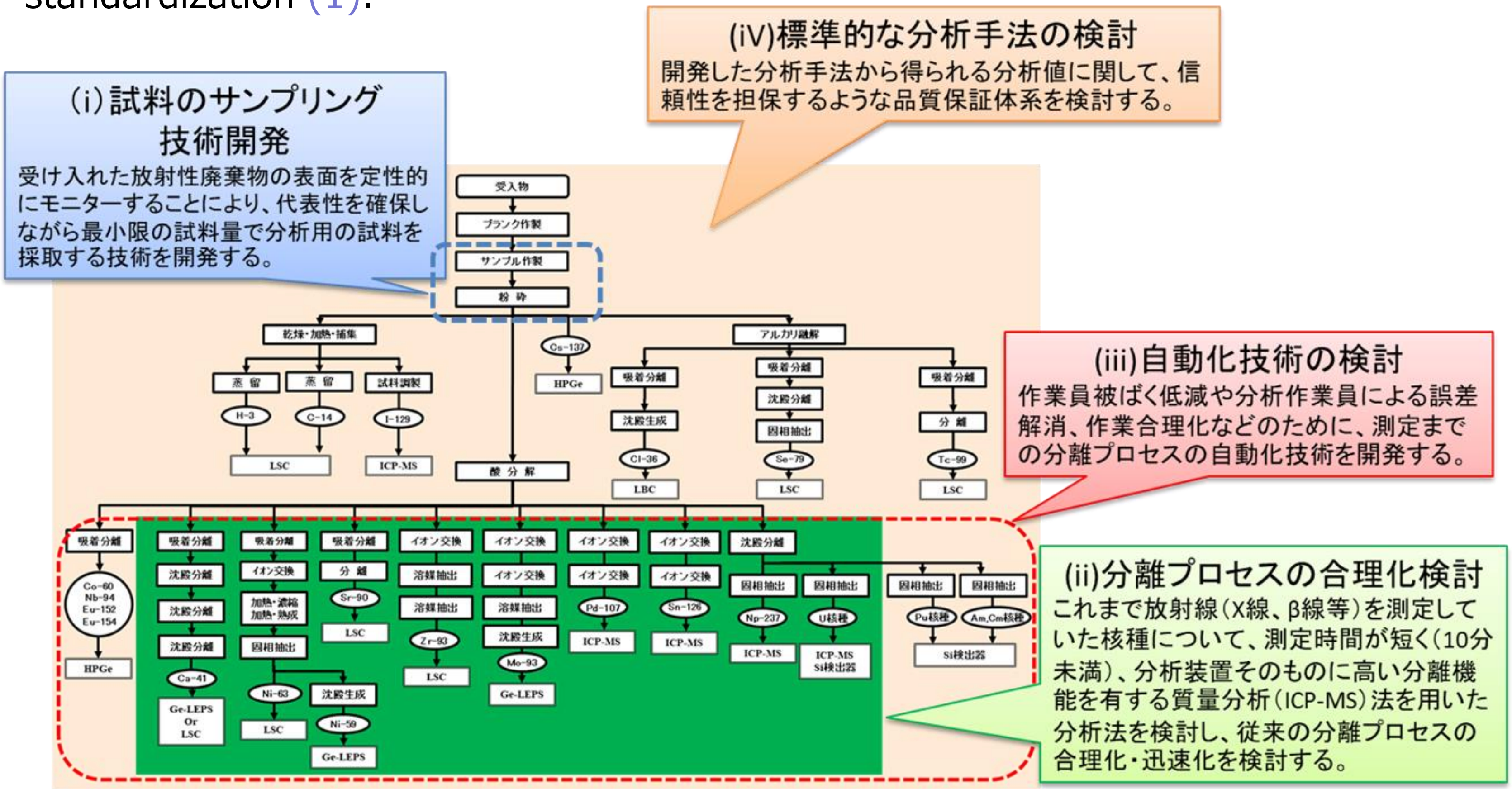

(1) Calculation model for estimation of various waste, which contaminated via waster and air.

(2) DQO process applied for waste management

*1 : https://irid.or.jp/wp-content/uploads/2021/12/2020010kotaihaikibuturev2.pdf
*2 : https://irid.or.jp/wp-content/uploads/2021/01/2019011kotaihaikibutsu_02.pdf

# II. Analysis of radioactive waste Analysis methods for DTM nuclides

- Various radioactive nuclides including ”Difficult-To-Measure” nuclides, which are important for disposal safety are analyzed. Even gamma emitters, they need chemical separation before gamma spectra determination; revealing most radionuclides are regarded as DTM nuclides.
- To aim analysis method of efficient, less time and effort, R&D has been conducted; ICP-MS/MS application, automated chemical separation, sample distribution, standardization (1).



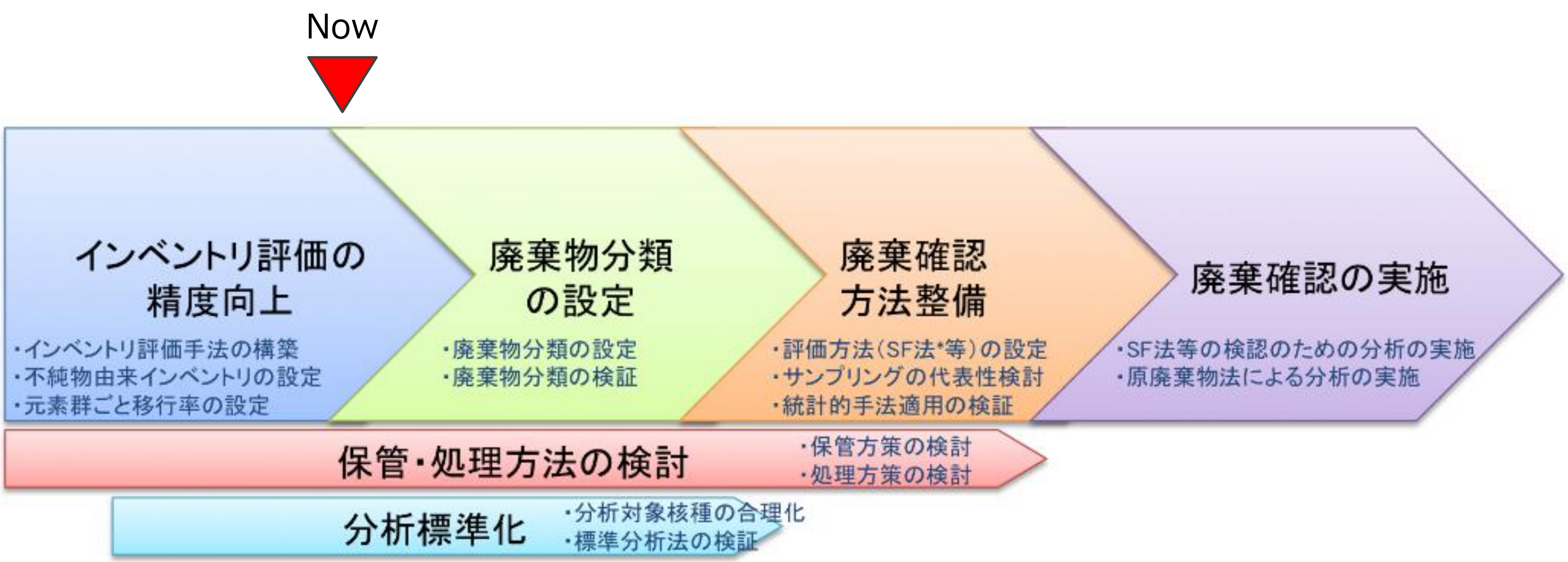

(1) Consolidated analysis method for waste sample and further improvement[*1]



*1 : https://irid.or.jp/wp-content/uploads/2021/12/2020010kotaihaikibuturev2.pdf

# II. Analysis of radioactive waste Aiming at disposal classification

- Objective of analysis is moved according to stepwise transition of waste management; improving inventory estimation, classification of waste, establishing waste confirmation method and operation of waste confirmation. (1)
- For these years, inventory estimation was major target along with analysis for establishing safe storage .
- Later, target will move to waste classification which considers future conditioning and disposal. Progress will be changed for waste due to difficulty of sampling and status of generation.




(1) 廃炉工程の進展に伴う分析目的の推移

# III. Fuel debris analysis - Required information and analysis technology -

## Information and analysis items necessary for safety and risk assessment

### A Safety / risk assessment through fuel debris removal to storage / management

<Retrieval, storage>

**Radiation exposure, scattering (α dust, FP, gas generation), criticality, heat generation**

**Safety / risk assessment items**

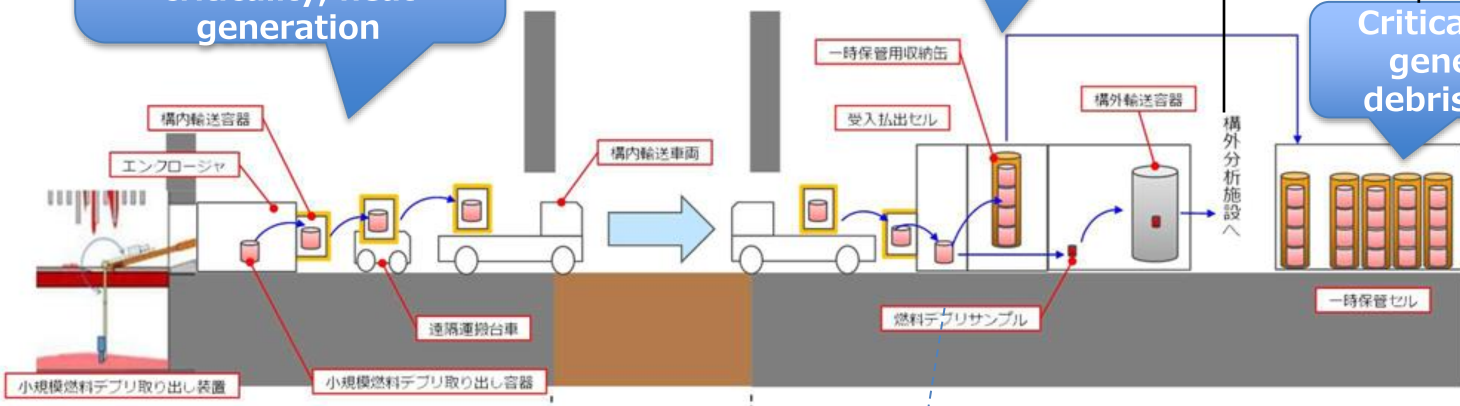
< Transfer, storage >

**Radiation exposure, scattering (α dust, FP, gas generation), criticality, heat generation**

< Temporary storage >

**Criticality, heat generation, debris amount**

Analytical facility (Okuma, Ibaraki area)





**Analytical needs**

- Appearance and weight
- Surface dose rate

- Appearance and weight
- Surface dose rate
- Non-destructive analysis of nuclear fuel materials, etc.(Sorting with radioactive waste) *

*) has not been established

Example on P.17

- Surface dose rate, dimensions (particle size), density, hardness・ Nuclide inventory, composition・ Chemical form, surface condition・ Moisture content, organic matter content, hydrogen・ FP emission behavior, thermal characteristics

Example on P.15,16

Feedback

Feedback

**Analysis / evaluation results**

## Information and analysis items necessary for safety and risk assessment

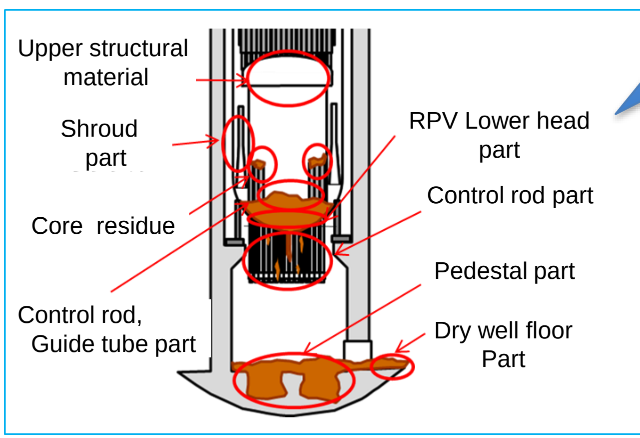
### B Safety / risk assessment in fuel debris processing

・In addition to the items applicable to temporary storage, the same analysis items as radioactive waste are applied. (Inventory evaluation: 38 Nuclide analysis, etc. )

### C Safety / risk assessment of fuel debris remaining in the container

**Current status**

Fig. Core status
(Estimated debris distribution)




**Safety / risk assessment items**

**Heat generation, criticality, scattering of fine particles, leakage of radioactive substances, (hydrogen generation)**

**Analytical needs**

・Surface dose rate, dimensions (particle size), density, hardness
・Nuclide inventory, composition
・Chemical form, surface condition
・Moisture content, organic matter content, hydrogen
・FP emission behavior, thermal characteristics

**Estimation**

・Plant information
・Internal investigation results
(Dosimetry, analysis of sediments, etc.)
・Analysis by calculation code such as thermal behavior
・Analysis of retrieved fuel debris
Analytical facility (Okuma, Ibaraki area)

**Analysis / evaluation results**

Feedback

Example on P.14

# III. Fuel debris analysis - Construction of a system to integrate data and information -

## 【JAEA Report】

This report summarizes the knowledge and experience of technology development implemented by JAEA, and systematically summarizes needs, issues, and analysis methods.

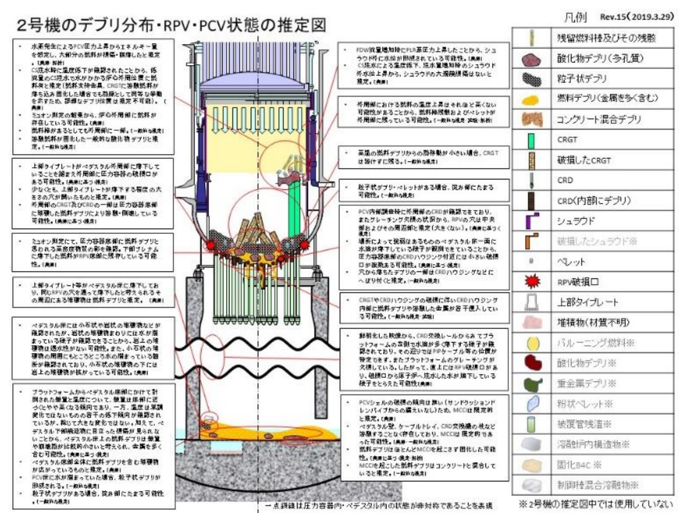

1. Introduction
2. Issues and analysis of fuel debris
   - Retrieval
     - Criticality control
     - Alpha-emitting dust, Pu/Am/residual FP during fuel debris cutting
     - Safety treatment, radiation dose
     - Heat generation and cooling measures
     - Hydrogen generation measures
     - RPV status
     - Index (burnup) for fuel debris analysis
     - Other issues
   - Analysis that could contribute to discussion of safeguards
     - Nuclear material accountancy
   - Storage management
     - Criticality safe control
     - Nuclide and radioactivity
     - Chemical stability, degradation
     - Optimization of storage facility
   - Processing and disposal
     - Importance of analysis
     - Details of analysis
   - Ascertainment of the cause of the 1F accident
     - Core relocation progression inside/outside RPV
     - Source terms
3. Fuel debris analysis flow
   - Analysis items and flow
   - Analysis items and apparatus
   - Relation to the fuel debris-related issues

A table of contents covering everything from removal to disposal and accidents

## 【Decommissioning research database（debrisWiki）】

- A hub of information required by users
- Provide users with up-to-date, technically-reviewed, and correct information.
- Compilation of knowledge created by experts in each field.
- Used as a work tool for field workers.

Core status map (Example of Unit 2)



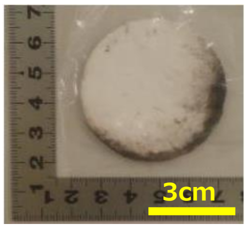

Same structure as the widely known Wikipedia

**Core status -** Estimated information on the distribution of debris throughout the reactor
**On-site information -** centralized search for 1F information
**Accident progression -** Based on the latest knowledge
**Analysis data -** Raw data, comprehensive interpretation of results
**Past knowledge -** Past SA accidents and 1F decommissioning

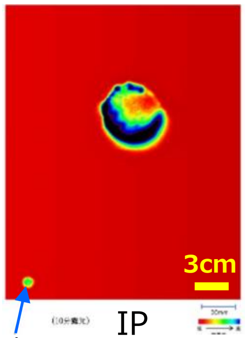
# III. Fuel debris analysis - 1F samples analysis (approach for samples of unknown composition) -

## Analysis technology and characterization (samples obtained from 1F internal investigation)

### 1/2u-SGTS plumbing smear (2020) [1]




Outer appearance

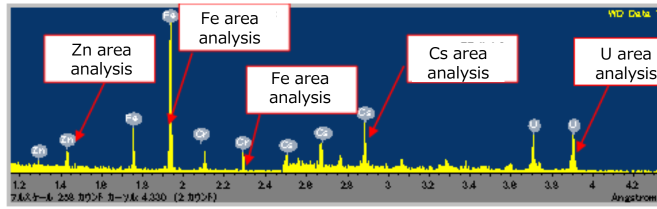


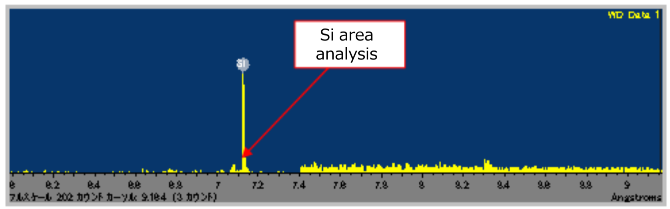

Std. radiation source (~48kBq)

IP (10min.)

### ◆ Outer appearance and IP

- Describe detailed information.
- Identify high-dose sites, collect samples.




### ◆ SEM/WDX

- Qualitative analysis, characteristic X-ray peak identification, characteristic X-ray image

Weight(ng)

| | XM20111 | XM20121 |
|---|---|---|
| Cr | $5.19\times10^2$ | $5.41\times10^2$ |
| Fe | $4.43\times10^4$ | $1.30\times10^5$ |
| Mo | $1.66\times10^1$ | $9.18\times10^2$ |
| U | $1.04\times10^1$ | $5.10\times10^0$ |

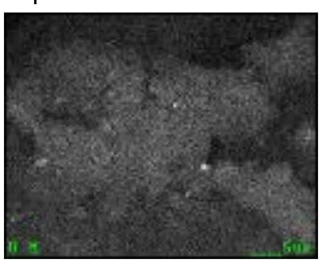
| | XM20111 | XM20121 |
|---|---|---|
| Cr | $1.2\times10^{-2}$ | $4.2\times10^{-3}$ |
| Fe | 1 | 1 |
| Mo | $3.8\times10^{-4}$ | $7.1\times10^{-3}$ |
| U | $2.3\times10^{-4}$ | $3.9\times10^{-5}$ |

U isotopic ratio

| | XM20111 | XM20121 |
|---|---|---|
| U235/U238 | $1.65\times10^{-2}$ | $1.9\times10^{-2}$ |
| U236/U238 | $2.53\times10^{-3}$ | $1.9\times10^{-3}$ |

### ◆ Dissolution／γ-ray／ICP-MS

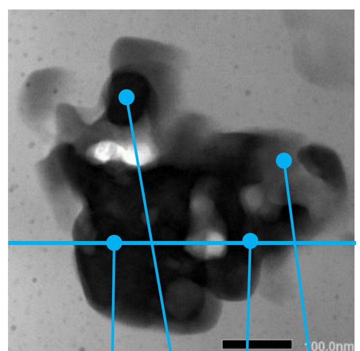
- Dissolution by 8N $HNO_3$ + HF, γ-ray of the residue, confirmed low $^{137}$Cs (less than 1/100)
- Quantitative analysis and isotope ratio evaluation (ICP-MS)
- Uncertainty evaluation



Elemental map of U
(Including influence of Ag signal)

### ◆ SEM/EDX

- Describe the basis for identification and overlap
- Describe the grounds for selecting the target area for TEM analysis




① ③ ② ④

U particle

### ◆ TEM

- Base of identification and the range of quantitative analysis.
- Increase the accuracy of oxygen analysis by ultra-thin film processing and zeta factor method.

Composition of main elements (at%)

| | O | Mg | Al | Si | Cr | Fe | Ni | Zn | Zr | Ag | U |
|---|---|---|---|---|---|---|---|---|---|---|---|
| ① | ~70 | 0.2 | 0.3 | 0.2 | 0.1 | 3.0 | 0.0 | 0.3 | 0.1 | 0.7 | 24.7 |
| ② | ~70 | 0.1 | 0.3 | 0.2 | 0.2 | 8.5 | 0.0 | 0.3 | 0.1 | 0.7 | 21.1 |
| ③ | ~70 | 0.0 | 0.4 | 0.0 | 0.1 | 3.0 | 0.0 | 0.2 | 0.1 | 0.7 | 23.1 |
| ④ | ~70 | 0.1 | 0.3 | 0.0 | 0.2 | 12.1 | 0.0 | 0.2 | 0.0 | 0.5 | 14.7 |

It is possible to estimate knowledge on the chemical properties of fuel debris and to estimate and evaluate accident scenarios, and provides basic knowledge for estimating the situation inside the reactor based on analysis.

https://irid.or.jp/wp-content/uploads/2021/09/2020004seijyouhaaku.pdf

# III. Fuel debris analysis - Analysis system and improving the quality of analysis data -

Technology and system for evaluating the characteristics of fuel debris will be established as an approach for samples with unknown composition.

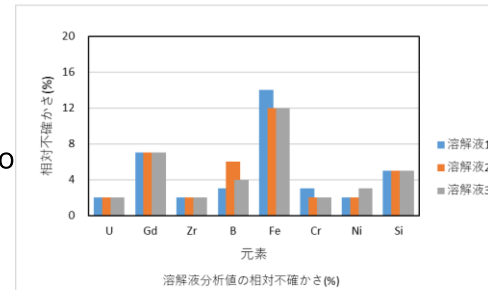
The simulated fuel debris prepared by an independent organization will be supplied to the hot laboratories in Japan. The elemental composition will be analyzed using the own technology possessed by each laboratories.

Simulated fuel debris (uniform composition)
・Including U
・1F composition

Tohoku University


| Hot laboratories | Dissolution method | Dissolution rate | Remarks |
|---|---|---|---|
| NDC | $HNO_3$ | ～60% | ・Simple method<br>・Dissolves U and B compounds<br>・Complement the overall elemental composition by analyzing residue |
| JAEA Oarai | $HNO_3$ + HF | ～90% | ・Improved dissolution rate by adding a small amount of HF |
| NFD | Aqua regia +HF | 98% | ・Almost complete dissolution<br>・High accuracy<br>・Possibility of fluoride precipitatio |
| JAEA NSRI | Alkali fusion | 100% | ・Complete dissolution<br>・High accuracy<br>・Contamination of alkaline reagent and crucible components |

Conducted by remote control within the cell




Verification of accuracy and error for each analysis target element and operation

Analysis techniques for 4 basic evaluation items (morphology of analysis samples, nuclide/element content, phase state/distribution, density, etc.), which are important for fuel debris analysis items, are defined and shared among the parties concerned.

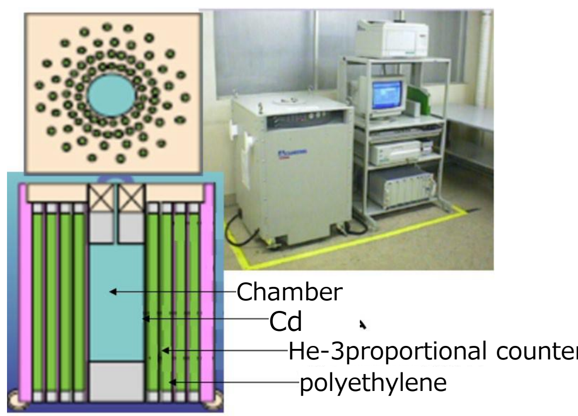
# III. Fuel debris analysis - Non-destructive assay technology for sorting of fuel debris and radioactive wastes -

- Destructive analysis of recovered PCV materials up to the order of kilograms stored is difficult.
- In order to reflect the sorting of fuel debris and radioactive waste in the future, non-destructive assay technology will be developed to sort them according to the amount of nuclear fuel material.

**Candidate methods currently under consideration in the IRID project:**
**(Participating organizations in the project: MHI, Toshiba ESS, Hitachi GE, JAEA collaboration)**
**(1) Active neutron method, (2) Passive neutron method, (3) Muon scattering method,**
**(4) X-ray CT method, (5) Passive gamma ray method**

A cooperative system consisting of 3 centers and 4 divisions has been established in JAEA, and preparations for testing (examination of test methods, production of simulated models, changes in authorization, etc.) are underway. Elemental technology verification tests for (1), (2), and (5) will mainly start in 2023.


Active neutron method (JAWAS-T)
NSRI/NUCEF/BECKY




Passive neutron method (PSMC)
NCL/Pu-center

Considered for use in passive gamma-ray measurement, etc.
NSRI/RFEF

Overview of basic test equipment for non-destructive assay measurement methods
(JAEA facilities and equipment scheduled to be implemented from 2023)

# IV. Technical issues and future perspective

## Continuing analysis and preparing for next step

→ Analysis will conducted at existing laboratories in Ibaraki-area. Newly built Laboratory-1 at Fukushima Daiichi will start operation near future.

→ Object should be determined for each waste. Obtained data will be utilized for establishing inventory estimation and be constituent knowledge base, which is essential to disposal safety.

## Analysis method development

→ R&Ds for improvement and DTM nuclides and sentencing waste and fuel debris will be conducted.

## Human resource development and international cooperation

→ Continuous human resource development is carried out through guidance by nuclear fuel and radiation experts and practical experience at facilities.

Develop JAEA staff and external human resources (joint research, MEXT projects, etc.)

→ Collaboration with overseas related organizations will be promoted through OECD/NEA projects and joint research.

# V. Conclusions

- JAEA will continue to support the analysis and sampling work at the 1F decommissioning in order to build knowledge and technology that can conduct safety and risk assessment as an all-Japan effort with NDF and TEPCO.

- Analysis of radioactive waste will soon begin at the Laboratory-1 of Okuma Analysis and Research Center. Efforts will be made to reduce risks in waste management and develop waste classifications for waste disposal. Analysis of the existing facilities will also continue to contribute to the steady progress of 1F decommissioning.

- Preparation for fuel debris analysis and development of a decommissioning research fundamental database that can be used by workers and users are in progress.

- We will utilize the current research and development for human resource development toward the construction and operation of the Okuma Analysis and Research Center and the TEPCO HD Comprehensive Analysis Facility.