Basics for Sampling and Analysis at Nuclear Facilities

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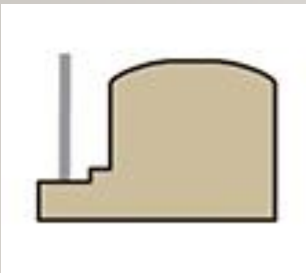
- Analysis at Nuclear facilities
- Classification and activity contents of radioactive waste
- Characteristics of waste in Fukushima Daiichi NPP
- General flow in waste analysis
- Requirements for waste analysis
- Finally

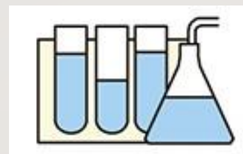
ANALYSIS AT NUCLEAR FACILITIES

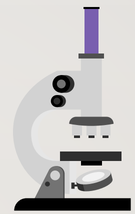
Analysis: the act of examining a substance, especially by separating it into its parts, in order to discover what it is or contains (Cambridge dictionary)



Operation of nuclear facilities

Nuclear waste generation



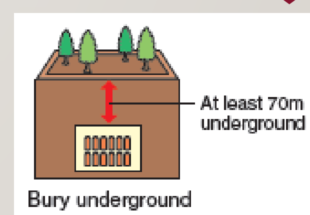


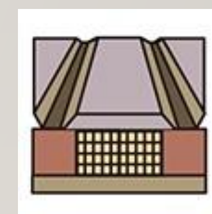
Analysis

- Nuclides ^{90}Sr, ^{241}Am, etc.
- Activity (Bq)
- Distribution (homo- or heterogeneous)
- Chemistry (crystal structure, solubility, stability, etc.)

Action Planning Based on the Analysis results

- Selection of safer storage methods
- Waste processing and conditioning plan
- Waste disposal plan
- Waste management cost estimation

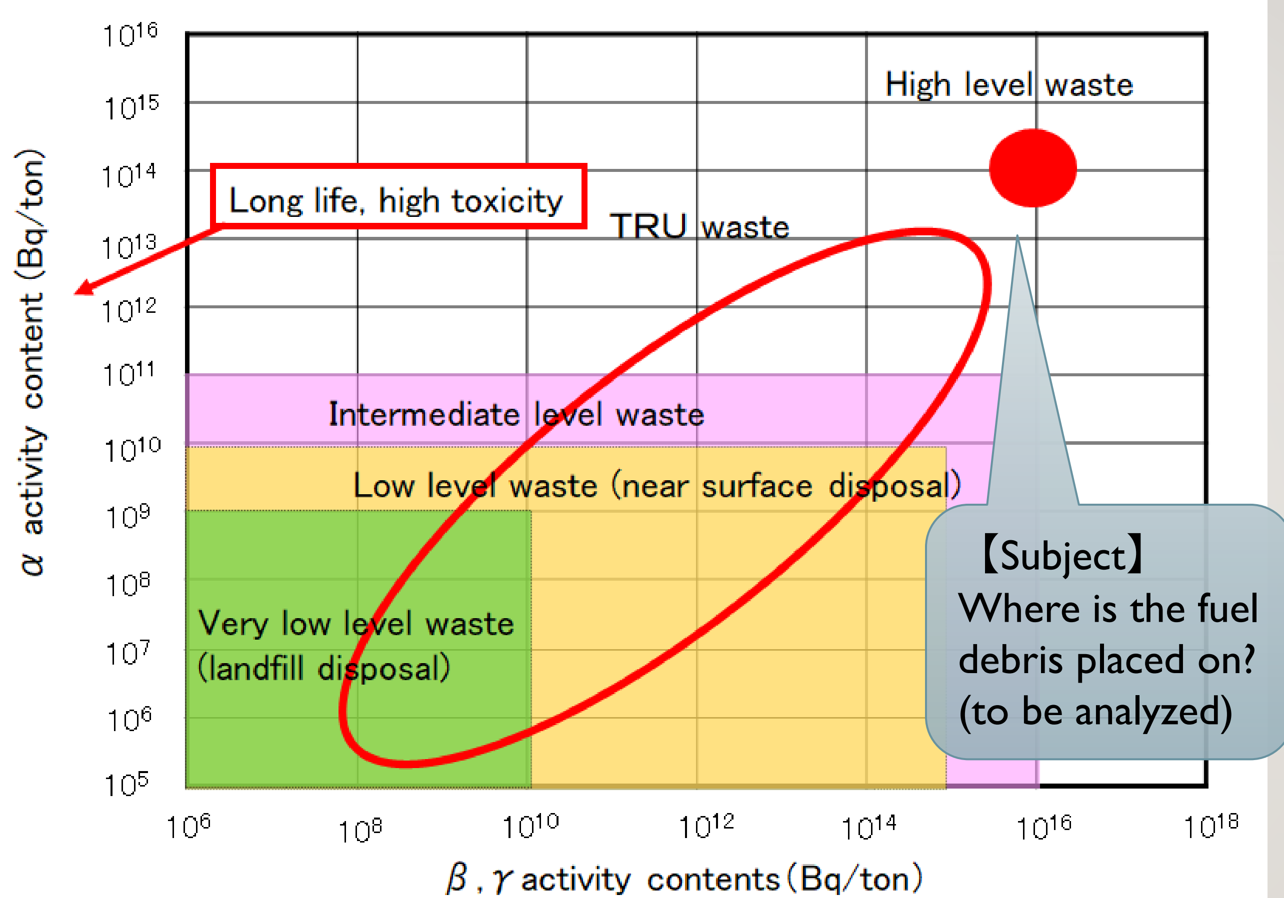




Future Disposal

!) If analysis data is insufficient, more conservative and over equipped storage and disposal facilities could be selected. A lack of analysis data could result in larger waste volume.

Classification and activity contents of Radioactive waste



- A suitable disposal method is selected depending on the activity contents and origin of the waste.
- Treatment and conditioning will be conducted for safe disposal facility operation.

For adequate radioactive waste management at FDNPP, the "Analysis" is critically important. That should cover not only activity contents, but also hardness, heat, chemical feature, and etc.

Quantity and Types of Waste Generated During Decommissioning

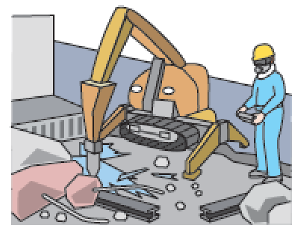
The total amount of waste generated in decommissioning a 1.1 million kW class boiling water reactor (BWR) is approx. 536,000 tons.

Decommissioning of normal reactors

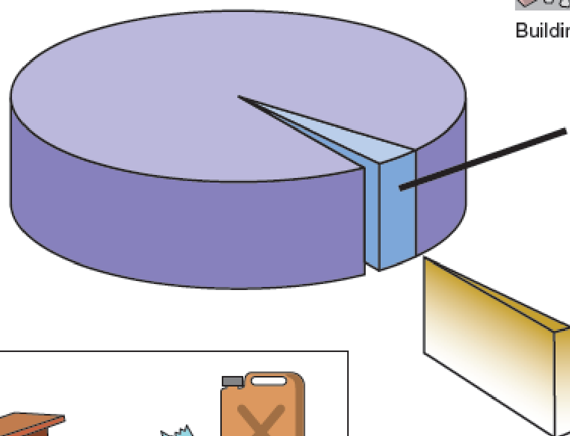
Waste that does not emit radioactivity

Approx. 93%

(Mostly waste concrete: approx. 495,000 tons)



Building concrete, glass, metal, etc.

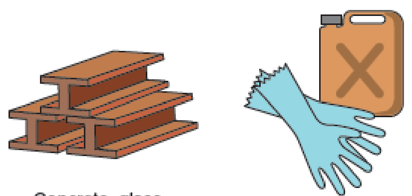


Materials below clearance level

Approx. 5%

(Metal and concrete waste: approx. 28,000 tons)

An appropriate waste classification is demanded to perform this ideal decommissioning.



Concrete, glass, metal, etc.

Materials used in dismantling

Low-level radioactive waste **Approx. 2%**

(Mostly metal waste: approx. 13,000 tons)

Source: Nuclear and Industrial Safety Agency (at the time), Decommissioning of Nuclear Facilities

Cost estimation codes for the decommissioning of normal nuclear facilities are proposed. These methods allows easy estimation of decommissioning costs based on the characteristics and similarities of facilities, dismantling methods, etc.

From JAEA R&D Review 2020-21

Research and Development on Fuel Reprocessing, Decommissioning, and Radioactive Waste Management

8-1 Toward Public Use of the Decommissioning Cost Estimation Code DECOST

—Development of a Manual for the DECOST—

Table 8-1 Input datasheet example for estimating the dismantling cost using DECOST

The information required for estimating the dismantling cost includes the unit cost per worker, building information related tc of the facility, special equipment information for individually evaluating the work required, and waste-related information fo total amount of work. In the manual, methods to obtain this information are explained and a summary datasheet is pro necessary information acquired from the Japan Power Demonstration Reactor (JPDR) is summarized. JPDR was the first to carry out nuclear power generation and was the first facility to demonstrate that we could dismantle and demolish a r

	Input information	Data			Input information	Data			Input information	
	Type of facility	Reactor		Special equip.	Accelerator shielding (Metal)	0	t	Waste related info.	Concrete_CL	
	Consumption tax	3	%		Accelerator shielding (Concrete) by wire saw	0	t		Concrete_NR	
Unit cost	Worker	XXXX	¥/man-day		Accelerator shielding (Concrete) by batch removal	0	t		Miscellaneous solid_L0	
	Manager of radiation management	XXXX	¥/man-day		Remote dismantling (Cell)	0 ¥ 0	n		Miscellaneous solid_L1	
	Manager of work management	XXXX	¥/man-day	Waste related info.	Metal_L0	0	t		Miscellaneous solid_L2	
Building info.	Decontamination systems	0	n		Metal_L1	44	t		Miscellaneous solid_L3	
	Floor area of cell	0	m²		Metal_L2	118	t		Miscellaneous solid_CL	
	Floor area of controlled area	23800	m²		Metal_L3	78	t		Miscellaneous solid_NR	
	Floor area of building (steel slate)	0	m²		Metal_CL	865	t		Casks	X
	Safe-storage period	0	year		Metal_NR	1324	t		1m³ steel containers	X
Special equip.	Lining weight	23	t		Concrete_L0	0	t		Drum (epoxy coatings)	X
	Centrifuge weight	0	t		Concrete_L1	60	t		Drum (galvanized containers)	X
	Large sized GB weight	0	t		Concrete_L2	83	t		Drum (concrete linings)	X
	Small sized GB weight	0	t		Concrete_L3	1477	t		Flexible container	X

JAEA-Testing
2018-002
DOI:10.11484/jaea-testing-2018-002

原子力施設廃止措置費用簡易評価コード
（DECOST）利用マニュアル

The User Manual of the Simplified Decommissioning Cost Estimation Code for Nuclear Facilities (DECOST)

高橋 信雄　末金 百合花　阪場 亮祐　黒澤 卓也
佐藤 公一　目黒 義弘

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核燃料・バックエンド研究開発部門
廃止措置技術開発室

Decommissioning Research and Development Office
Sector of Nuclear Fuel, Decommissioning and Waste Management Technology Development

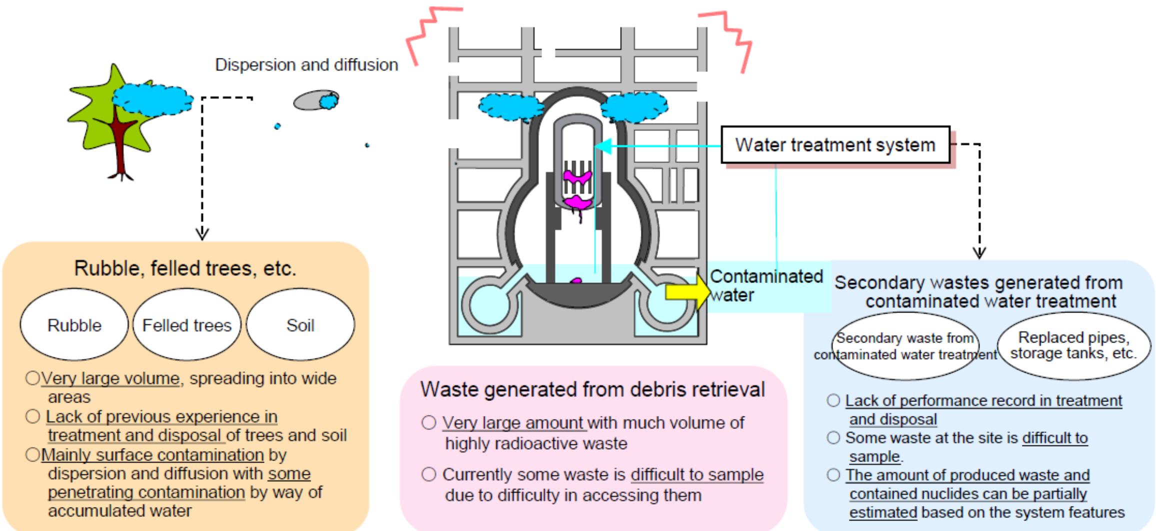
JAEA-Testing

Required info.: The total floor area of the controlled area and the amount of dismantled waste, classified by disposal type (e.g., radioactive or non-radioactive) and by materials (e.g., concrete or metal), etc.

WASTE IN FUKUSHIMA DAIICHI NPP……….

Characteristics of Waste Generated by the Fukushima Daiichi Accident (Estimation)

- Waste generated out of control due to the accident
- Contamination originated from nuclear fuel in the reactor core of Unit 1 to Unit 3*
- Difficulty in estimating the amount of waste produced with the varying status of decommissioning work
- Extremely limited data due to an extensive contamination area and high-radiation locations (particularly for the composition of nuclides with long half-life)



From TEPCO holdings inc.

From IRID Subsidy Project of Decommissioning and Contaminated Water Management in the FY2016 and FY2017 Supplementary Budgets, "R&D for Treatment and Disposal of Solid Radioactive Waste ", Accomplishment Report for FY2018, July 2019

- Difficulty in estimating the amount of waste produced (Necessity of Analysis) .
- The amount of waste will change depending on the way of decommissioning mission.
- Limited analysis data points due to the limitation in the hard- and software.

Challenge: Development of the Analysis strategy covering both hardware and HR.

General flow in waste analysis 1. Planning and sampling

Sampling at Fukushima Daiichi NPP

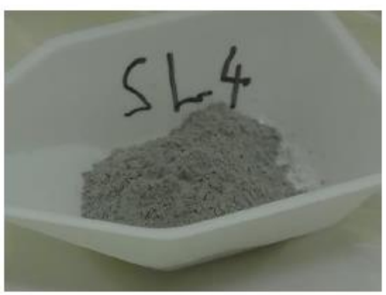
https://www.jaea.go.jp/04/ntokai/fukushima/fukushima_02.html

- Set the target of the analysis. (What is the purpose of this analysis?)
- Make a suitable plan of the sampling. (sample numbers and its amount)
- Select sampling techniques. (remote or contact sampling)
- Check the achievability of the plan in the point of site situation.

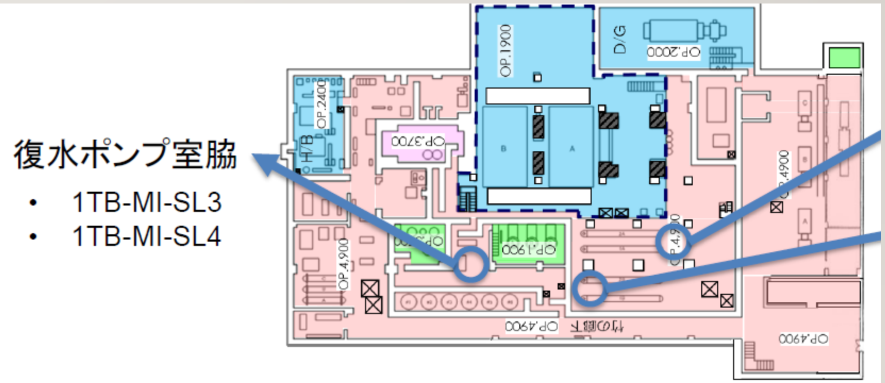
Sludge sampling at the turbine building

出典：JAEA-IRID　廃炉・汚染水対策チーム会合／事務局会議（第28回）, 平成28年3月31日

1TB-MI-SL3



1TB-MI-SL4



1号機タービン建屋地下1階 ※1

General flow in waste analysis 2. Pretreatment

High dose ← Low dose

Operability : Low → High

Concrete Cell with manipulators

図６　アルゴン循環型グローブボックス
[出典]（独）日本原子力研究開発機構：パンフレット「燃料サイクル安全工学研究施設NUCEF」、原子力科学研究所 安全試験施設管理部（2006）、p.10

Globe Box

Fume Hood (Draft chamber)

- Conditioning and adjustment of the samples for the following analysis.
- Select suitable hardware depending on the dose level and feature of the sample.
- Generally, the operability of high shielding hardware is low. (trade-off relation)

Photos from JAEA web https://www.jaea.go.jp/04/ntokai/hot/hot_04.html and ATOMICA


MB

General flow in waste analysis 2. Pretreatment (in chemistry)

Liquid samples containing particles

Filtration to separate particles from liquid

Particles dissolution

Dissolution by heated and concentrated nitric acid

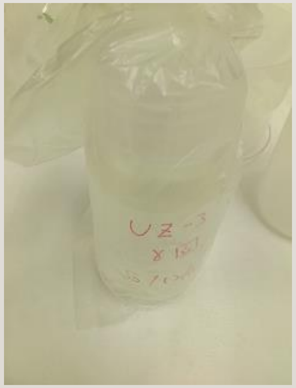
Ion exchange to reduce salinity

dillution

Liquid sample

Liquid phase

dillution



Liquid sample

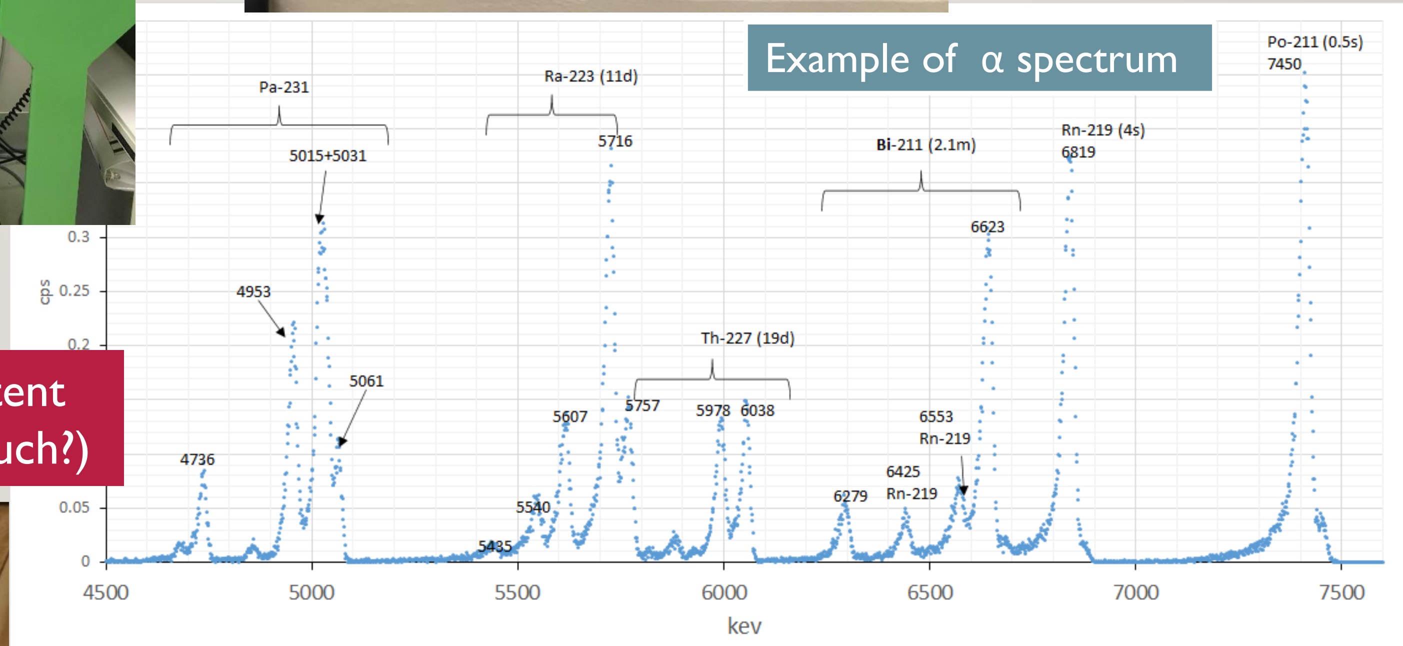
Radiation counting
- γ-ray spectrometry
- α spectrometry

General flow in waste analysis 3.Radiation counting (activity)

Ge semiconductor detector (γ ray spectrometry)

Si semiconductor detector (α spectrometry)

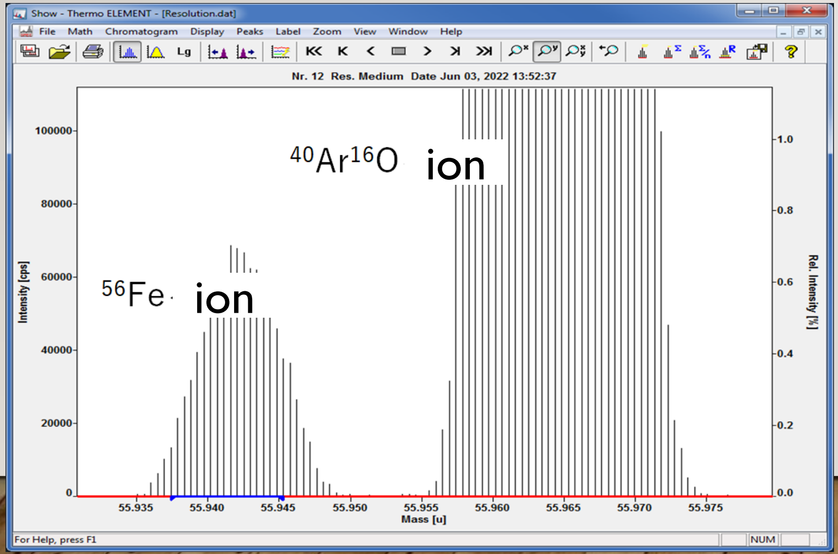
Example of α spectrum



Determine activity content (which nuclide?, how much?)

General flow in waste analysis 4. Mass spectrometry (activity det.)

Photo：High Resolution ICP-MS system ”Thermo Fisher Scientific ELEMENT 2” @IMRAM Tohoku Univ.



Ionize elements in the liquid sample by an inductively coupled plasma. The atomized elements are detected by a mass spectrometer. Modern powerful tool for several radioactive nuclides.

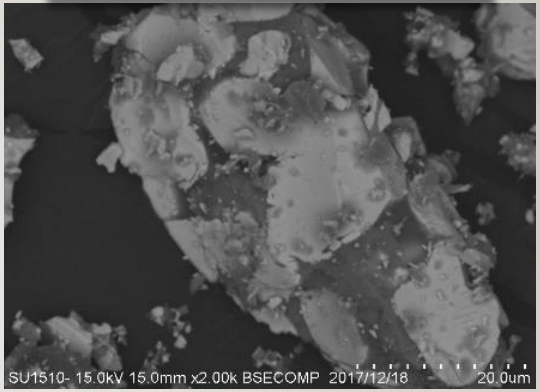
General flow in waste analysis
5. Instrumental analysis (characterization)

Sample
(simulated fuel debris)

Investigate the chemical and physical features of the target.

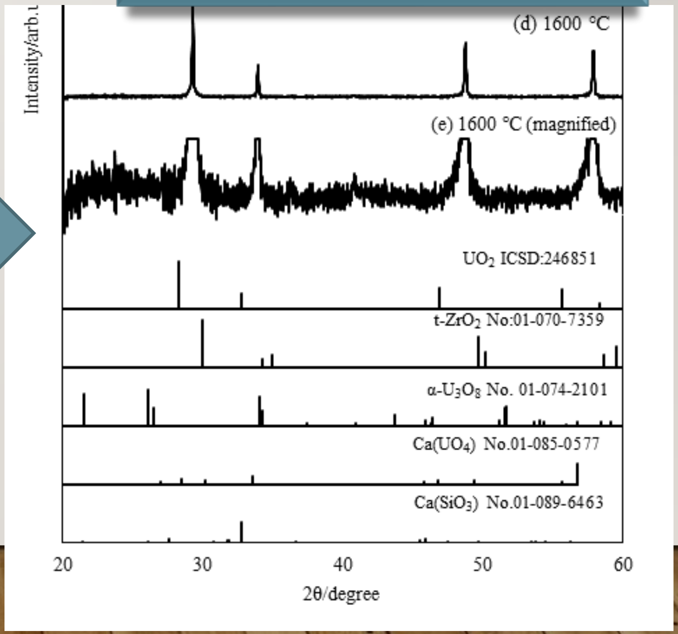
Scanning Electron Microscope(SEM)

Analyze fine structure



Powder X-ray diffract meter (p-XRD)

Analyze crystal structure



A. Kirishima et al. / Journal of Nuclear Materials 527 (2019) 151795

デブリとみられる堆積物が大量に付着した福島第1原発2号機の原子炉格納容器底部（東京電力提供）

福島第1デブリ　固溶体化で化学的安定

東北大など　長期保管へ知見

東京電力福島第1原発事故で溶け落ちた核燃料（デブリ）の化学的性質を巡り、複数の物質が混じり合い均一の状態になる「固溶体化」という現象が起こると化学的により安定し、放射性物質が水に溶け出しにくくなると、東北大と日本原子力研究開発機構、京都大の研究グループが発表した。デブリの保管や処理を今後検討する際の重要な知見になるという。

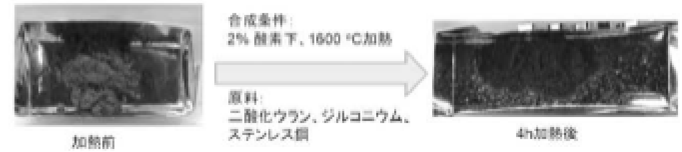
研究グループはデブリの性質を把握するため、ウランと燃料被覆管に使われるジルコニウム、原子炉の材料であるステンレス鋼を1600度に加熱し、模擬デブリを合成した。

解析の結果、ジルコニウムや、ステンレス鋼の主成分である鉄がウラン結晶中に溶け込む固溶体化が起きたことが判明。純水や海水に最長400日間浸したが結晶構造に変化は見られず、元のウランよりも化学的に安定することが分かった。

さらに、デブリに含まれるとされる毒性の強いネプツニウムやアメリシウムを模擬デブリに添加して純水や海水で試験したところ、溶け出した放射性物質の割合は0・08％未満と非常に微量だった。

東北大多元物質科学研究所の桐島陽教授（放射化学）は「デブリの長期保管や処理を考える上で、固溶体化が重要な鍵を握ることが分かった」と語る。

河北新報　朝刊3面
2022/06/20



合成した模擬デブリの一例。加熱前は茶色の粉末だったが、加熱後は一部が溶けて固まったような黒い塊状になった（東北大提供）

燃料デブリ　固溶体化で安定

東北大など　保管・処理法検討に一役

東北大学多元物質科学研究所の桐島陽教授らは、東京電力福島第一原子力発電所の事故で核燃料が溶け落ちた燃料デブリは、配管などに含まれる金属などが核燃料成分中に溶け込んで固体となる「固溶体化」が起こることで、元の状態より化学的に安定化することを示した。固溶体化すると、水や海水に漬かっても変化しにくく、放射能毒性の強いアクチノイドが溶け出しにくくなる。取り出し後の燃料デブリの保管や処理方法の検討に役立つ。日本原子力研究開発機構、京都大学との共同研究。

核燃料主成分の二酸化ウランと、被覆管材料に含まれるジルコニウムや炉内構造材に使われるステンレス鋼などを1600度Cに加熱して模擬デブリを合成し、その性質を調べた。

酸素有無や原料の組み合わせなどを変えた37種の模擬デブリを分析した結果、ステンレス鋼中の鉄やジルコニウムが高温で反応すると、二酸化ウランに鉄やジルコニウムが溶け込んだ固溶体を形成することが分かった。

この固溶体について水中での経年変化を観察した結果、どの模擬デブリも純水および海水中で400日間、結晶構造などに変化がなく、非常に安定だと分かった。

さらに、アクチノイド元素のネプツニウムやアメリシウムを添加した模擬デブリを作って水中での溶け出しやすさを調べると、純水、海水ともに溶け出す割合は0・08％未満と微量だった。固溶体化が進むと、さらに安定性が高まった。

骨との結合強度3倍

阪大など　チタン合金製脊椎ケージ

大阪大学の中野貴由教授らは国立病院機構北海道医療センターの伊東学統括診療部長、帝人ナカシマメディカル（岡山市東区）と共同でチタン合金製の脊椎ケージを開発した。レーザーで粉末を選択的に溶かして固めた層を積層するレーザー粉末床溶融結合法という金属3Dプリンティング技術をもとに「ハニカムツリー構造」と名

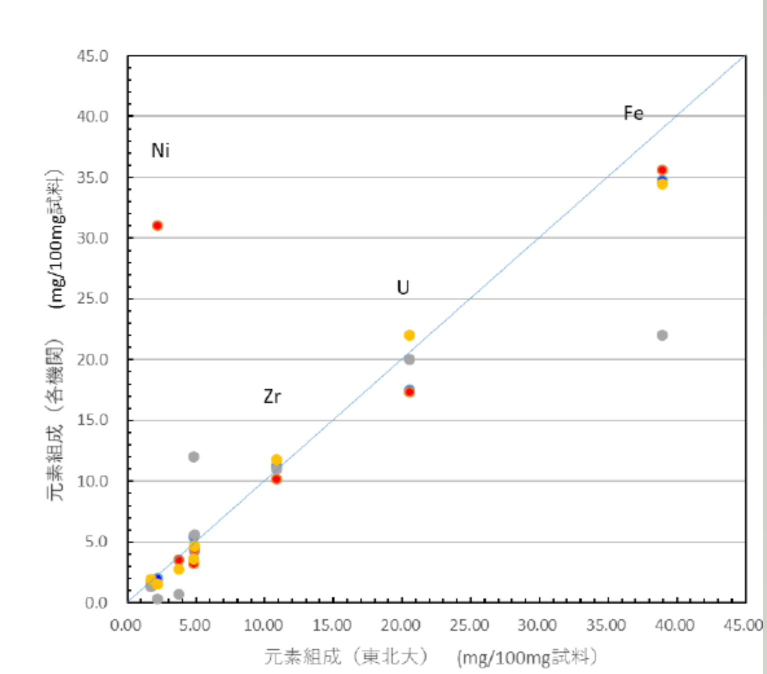
日刊工業新聞　21面
2022/06/22

General flow in waste analysis
6. Compilation and interpretation of analysis results

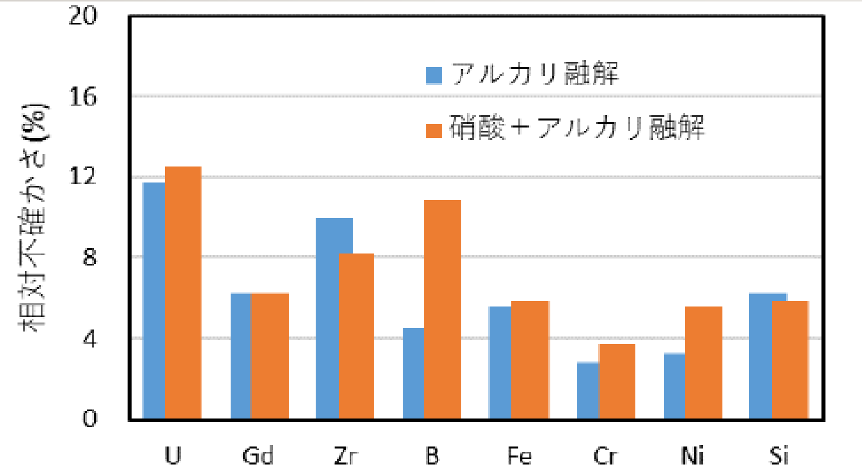
不溶解性残渣成分を合算*した評価値（U模擬燃料デブリ試料）

元素名	含有量（mg/100mg）					
	東北大	NDC	JAEA大洗研	NFD	JAEA原科研	
	【基準値】	硝酸溶解	硝酸溶解	王水・フッ酸溶解	硝酸溶解＋残渣アルカリ融解	全量アルカリ融解
U	20.5 ±0.1	20.0 ±0.6	21.9 ±1.0	17.5 ±0.2	16.5 ±0.8	17.3 ±2.0
Gd	1.7	1.3 ±0.2	1.9 ±0.1	1.5 ±0.2	1.56 ±0.11	1.59 ±0.08
Zr	10.8	11 ±1.4	12 ±1.7	11.3 ±1.1	8.48 ±0.71	10.2 ±0.5
B	4.9	5.6 ±0.5	4.7 ±0.2	4.3 ±0.2	4.63 ±0.50	4.44 ±0.20
Fe	39.0	22.0 ±4.8	34.6 ±3.9	34.7 ±0.4	33.8 ±1.6	35.6 ±1.6
Cr	3.7	0.7 ±0.9	2.8 ±0.6	3.5 ±0.1	2.75 ±0.10	3.57 ±0.15
Ni	2.2	0.3 ±0.4	1.6 ±0.3	2.0 ±0.1	180 ±10	31.0 ±1.4
Si	4.9	12.0 ±2.4	3.6 ±4.5	5.4 ±0.4	3.40 ±0.17	3.22 ±0.16
O	12.2 ±0.1	-	-	-	-	-
不溶解性残渣	-	あり（溶解率60%）	あり（溶解率91%）	あり（溶解率98%）	なし	なし

注）不溶解性残渣が生じた場合は、溶解成分に残渣成分のEDS半定量値を加えた評価値を示す。



元素組成（最終評価値）の比較（0～45mg/100mg範囲）



- Evaluate and validate the data from each analysis step.
- Make a total interpretation of the analysis with multidimensional view. Then compile into a conclusive "fact of the sample".

Data from：【JAEA】令和2年度補助事業：燃料デブリの分析精度の向上及び熱挙動の推定のための技術開発の成果の概要　https://dccc-program.jp/3797

Requirements for waste analysis (in human resource aspect)

【General analysis flow】

1. Planning and sampling
2. Radiation counting (activity)
3. Mass spec. (activity)
4. Instrumental analysis (characterization)
5. Compilation and interpretation

【Personnel in analysis】

- Well trained in the operation of the responsible analysis procedure. Require abilities to detect trouble at the operation and to report it properly.

(**Skilled Technician)**

＊able to be educated in an equipped training facility

【Manager and Evaluator of the analysis】

- A person who well understands the objective of the analysis and is able to make a sampling plan.
- A person who is able to compile and validate variety of analysis data, then derives conclusion of the analysis project.

(Expert) How to educate? This education demands much time and cost. Need smart strategy!!

Finally

【Analysis of the radioactive waste generated in the decommissioning operation】

Purpose：Prepare a waste catalog that is necessary for the waste management.

Features：Difficulty in estimating the amount and character of waste.

Issues：**Limited analysis data due to the limitation in the hardware and personnel.**
Education of analysis managers and evaluators.

【Analysis of fuel debris】

Purpose① (decommissioning)

Collecting fundamental information (activity component, heat release, hydrogen generation, chemistry, etc.) for designing the safe storage, treatment process, and final disposal system of the debris.

Purpose② (Accident-progression analysis)

Acquiring information on what happened at the accident and how it progressed, which contributes the cause investigation and enhancement of the safety of nuclear facilities. More detailed information is demanded than “Purpose ①” .

Issues

Same ones as above mentioned. Additionally, more detailed analysis is required depending on the theme. As a result, purpose ② consumes more time and cost. Need smart arrangement between purpose ① and ②.