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Criticality Safety Control of Fuel Debris

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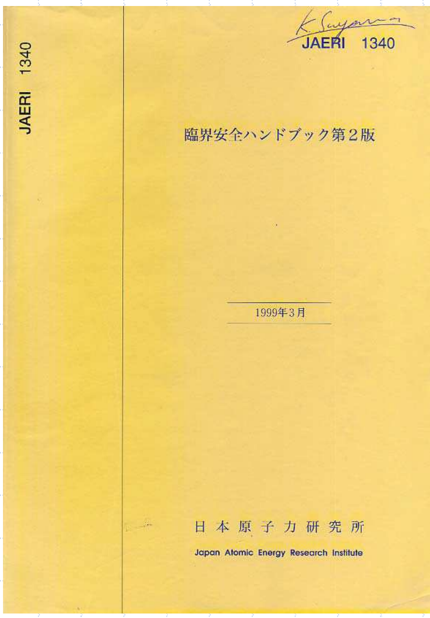
Criticality Safety – general issue

Criticality Safety



- Generally speaking, to prevent criticality, outside of nuclear reactors.
- Basic principle of criticality safety (Criticality safety handbook, *JAERI-1340*)
  - To prevent criticality for all situations that are technically convinced.
  - To have enough safety margin in each stage of design, fabrication, construction, and operation of facilities.
  - Need to consider kind(sort), amount, physical and chemical status of nuclear materials

How to keep enough safety margin

- Kind of nuclear materials
  - Which isotopes?
- Amount of nuclear materials
  - Nuclide density?

→ Controlled and Confirmed

- Consideration of physical and chemical status
  - Homogeneous/Heterogeneous(solution/array of rods)
  - uniform/un-uniform(concentration distribution)

Re-Criticality in Fukushima? – reported in 2011(1)

- From IAEA web site (https://www.iaea.org/newscenter/focus/fukushima/status-update):
  - On 1 November, TEPCO detected the possible presence of xenon-133 and xenon-135 gases sampled from inside the Primary Containment Vessel (PCV) of Fukushima Daiichi Unit 2. The presence of these short-lived radionuclides indicates that some nuclear fission may have recently occurred. TEPCO reported that no increases in radiation levels have been observed. According to TEPCO "even if a fission reaction is assumed to be on-going, its scale is extremely small and the reactor is in a stable condition as a whole."
  - TEPCO responded to this development by injecting 10 tonnes of boric acid solution (water containing 480 kg of boric acid) into the reactor from 02:48 to 03:47 local time on 2 November. Boric acid solution is used as a countermeasure to nuclear fission for its ability to absorb neutrons.
  - Further radionuclide analysis of the gas samples collected from Unit 2 is on-going and will be conducted in collaboration with the Japan Atomic Energy Agency (JAEA).

Re-Criticality in Fukushima? – reported in 2011(2)

➢ From IAEA web site (https://www.iaea.org/newscenter/focus/fukushima/status-update):

- Based on further analysis, Japanese authorities have concluded that the xenon concentrations are not due to a criticality event but rather from the spontaneous fission of curium-242 and 244. (Spontaneous fission is a form of radioactive decay that does not involve chain reactions and is characteristic of very heavy isotopes. Spontaneous fission occurs in low levels in all nuclear reactors.)

- This conclusion is based on three key factors outlined and discussed in the report:

- The inventory of Cm-242 and Cm-244 was calculated as was the concentration of Xe-135, resulting from the spontaneous fission of Cm-242 and Cm-244. If nuclear fission of the reactor's uranium fuel were occurring, at the lowest possible level, the levels of xenon detected would be several orders of magnitude higher than those measured. Current levels of xenon are consistent with those that would be generated by spontaneous fission of Cm-242 and Cm-244;

Re-Criticality in Fukushima? – reported in 2011(3)

➢ From IAEA web site (https://www.iaea.org/newscenter/focus/fukushima/status-update):

- If the core had been experiencing a criticality event, the injection of boron water should have stopped the criticality and terminated the generation of xenon. However, the xenon levels were not influenced by injection of boron water into the core; and

- If the core was undergoing a criticality event the temperature and pressure readings would be expected to rise as the event would increase heat production within the core. However, the temperature and pressure levels have not undergone any significant increases either before or after xenon were detected, indicating that no criticality event is occurring.

➢ No re-criticality event in Fukushima but very important issue.

Fuel Debris – What are difficult points?

Difficulty of Fuel Debris

- Kind of nuclear materials
  - which isotopes?
- Amount of nuclear materials
  - Nuclide density?

→ Not confirmed nor controlled after SA

- Consideration of physical and chemical status
  - Homogeneous/Heterogeneous(solution/array of rods)
  - uniform/un-uniform(concentration distribution)

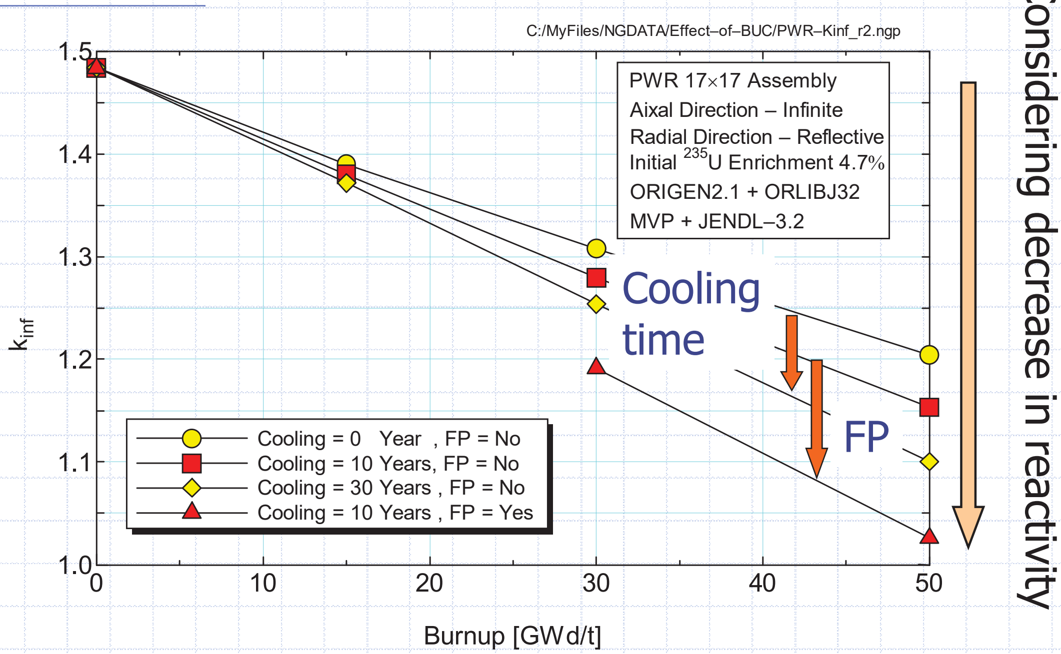
Reality

- We can say that the maximum ^{235}U enrichment is less than 5wt% (fabrication condition).
- When we use the condition "^{235}U enrichment is less than 5wt%";
  - The estimated lower-limit critical mass is almost 30kg for uniform U-H_2O system(JAEA-Data/Code 2009-010).
  - In case total amount of U is about 100t/reactor, we need ~3,333 units/reactor to contain fuel debris. (This does not include safety margin.)
  - Fuel debris is more than 100t because includes constructing materials.
- Is it possible to have additional conditions to control the criticality of debris to relax the situation?

We evaluate…(1)

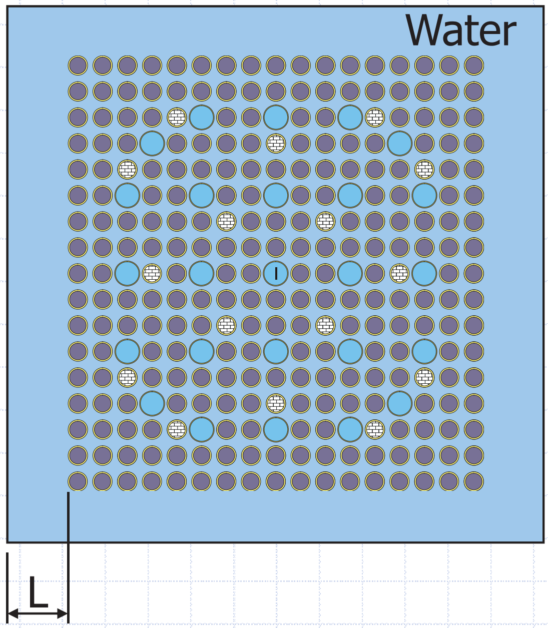
- Fuels are not fresh (average burnup ~ 20GWd/t).
  - "Averaged residue ^{235}U enrichment" is less than 5wt% because of burnup.
  - Accumulation of fission products.
  - By using burnup calculation codes, it is an easy task to evaluate the averaged residue ^{235}U enrichment.
- Burnup credit is an attractive option.
- However burnup credit is not adopted even in the case of the intact fuel transportation in Japan.

Example of change of k_{inf} during burnup



Evaluation of benefit of BUC (1)

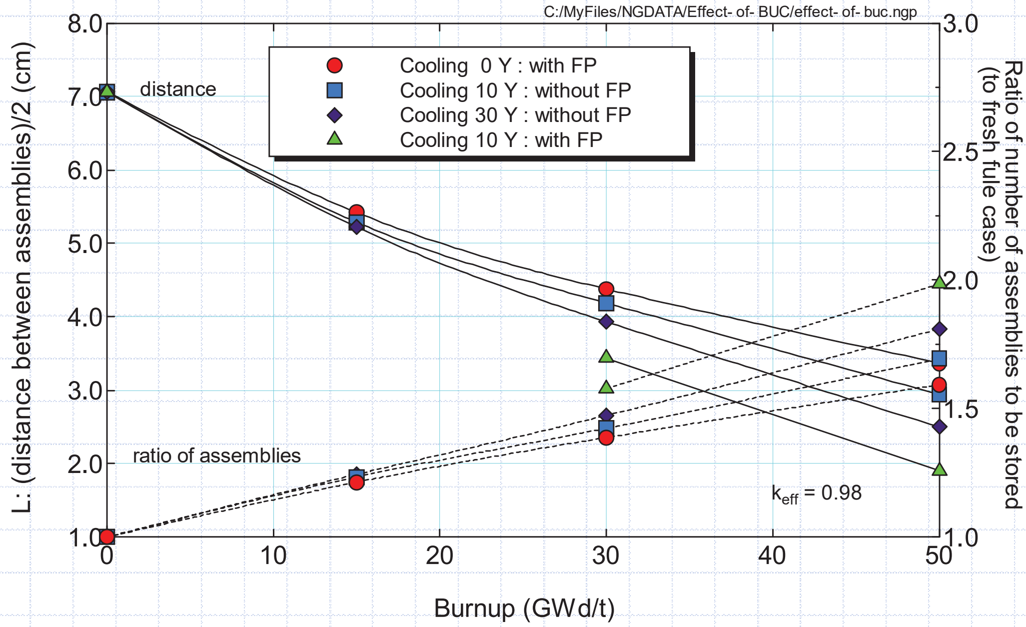
- Condition
  - PWR 17×17
  - ^{235}U;4.7wt%
  - ORIGEN2.1+ORLIBJ32
  - MVP+JENDL-3.2
  - Axially infinite、Radial Reflective
  - Water rods are replaced by UO_2rods.



To obtain L value of K_{inf} = 0.98

Evaluation of benefit of BUC (2)

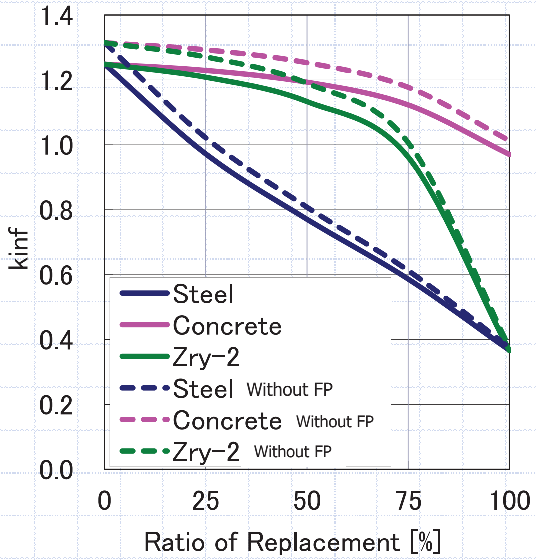
It is possible to store approximately doubled number of assemblies in unit surface under specific condition.



We evaluate…(2)

- Adding structure materials (Fe, Si, Zr etc) into the fuel debris increases the minimum criticality mass/volume/size of the fuel debris.
  - Fuel debris should contain them after SA.
- We can estimate the material property of the fuel debris by computer simulations or experiments.
- But no confirmation in Fukushima case.

Effect of Steel, Concrete, Zry-2



- Uniform UO_2-H_2O
- Uniform UO_2-H_2O+ Constructing Material
- Negative reactivity
  - Fe>Zry-2>Concrete

2013 AESJ Annual Meeting

We evaluate…(3)

- If the constituent elements of the control rods (Boron) exist in the fuel debris, the minimum criticality mass/volume/size should be increased.
- If residual gadolinium in the UO_2-Gd_2O_3 rods exists in the fuel debris, the minimum criticality mass/volume/size should be increased.
- During the process to compose the fuel debris, such materials may be well mixed in the fuel debris.
  - Born and gadolinium may have the same melting/mixing behavior with U.

- But no confirmation in Fukushima case.

Reality(1)

- The criticality safety control requires the confirmation/control/monitor of relevant parameters.
- Maximum/minimum of such parameters should be within a certain "limit", which could be controlled/confirmed.
- If we cannot confirm/control/monitor such parameters, these should be set at the upper/lower limit from the technical view point.

Reality(2)

- After the SA in Fukushima, people in this field quickly realized the necessity of the criticality safety control of the fuel debris especially when taking out it.
- Difficulty - this is because we have no procedures to confirm/control/monitor the relevant parameters.
- People would like to find possibility to relax tough parameters and conditions.
- But it is just the problem of the possibility. There is no definitive evidence nor confirmation procedure which could be adopted.
- We need further technical development.

Reality(3)

- Of course, criticality safety control is one of the key issues for decommissioning of Fukushima site considering the impact of the re-criticality on the society.
- We need clear strategy and consensus on this issue. Everything should be under controlled.
- Which parameters could be used to control the criticality of the fuel debris?

Technical Development for Criticality Safety of Fuel Debris in Japan – IRID

Fukushima - decommissioning

- Mid-long term road map towards the decommissioning (updated in June 2015).
  - The Council for the Decommissioning of TEPCO's Fukushima Daiichi Nuclear Power Plant
  - http://www.tepco.co.jp/en/decommision/planaction/index-e.html

- Removal of fuel assembly from SFP of 1F-1,2,3 – starting from FY2017
- Removal of fuel debris – starting from FY2021

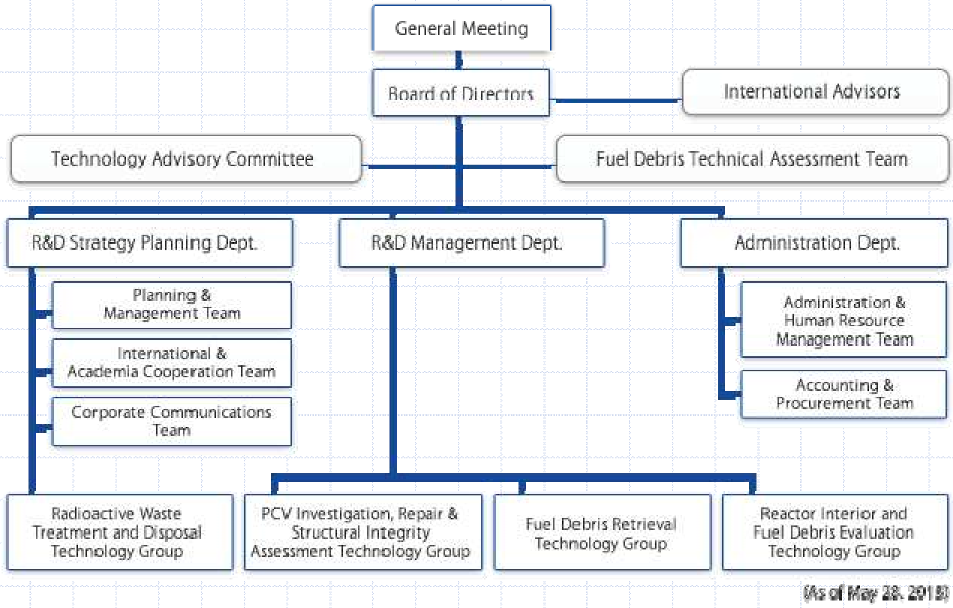
IRID - General information

- From August 2013, the technical development of decommissioning of Fukushima site has been carried out under the coordination of IRID (International Research Institute for Nuclear Decommissioning).

  - Composed of 18 institutes.(utilities, reactor vendors, JAEA, CRIEPI etc.)
  - To manage the funds and the technical program of the technology development for Fukushima decommissioning.

- http://www.meti.go.jp/earthquake/nuclear/pdf/140731/140731_01_041.pdf

IRID – Organization Framework



http://irid.or.jp/en/organization/

IRID for criticality safety(1)

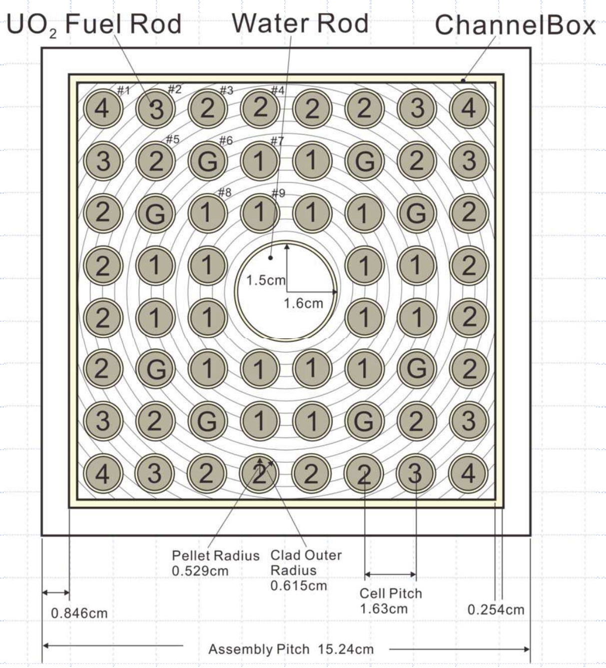
- Managing the funds from the Agency for Natural Resource and Energy.
- Three main vendors of NPP of Japan (Hitachi, Toshiba and Mitsubishi) have been carrying out the technical development for the criticality safety control of the Fukushima debris.
- JAEA also participates in the same program.

IRID for criticality safety(2)

- R&D for Criticality Control Technique
  - Evaluation of the Fuel Debris Criticality
  - Analysis Method for Power and FP Generation Behavior at the Criticality Accident
  - Sub-criticality Monitoring Technology for the Fuel Debris Removal
  - In-core Criticality Detection Technology
  - Criticality prevention technology
- See slides presented in Japan-Korea Summer School in August 2015

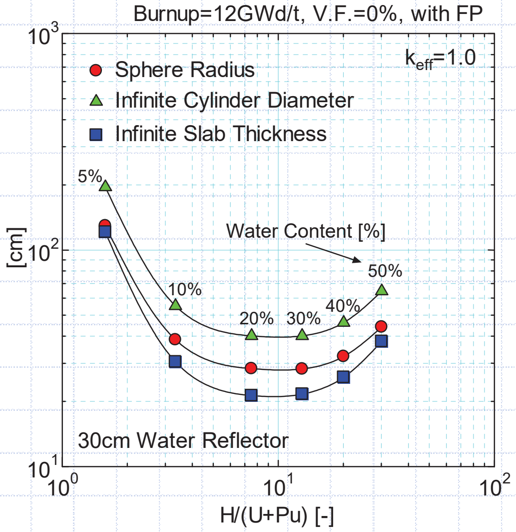
Technical Development for Criticality Safety of Fuel Debris in Japan – JAEA

Technical development in JAEA(1)

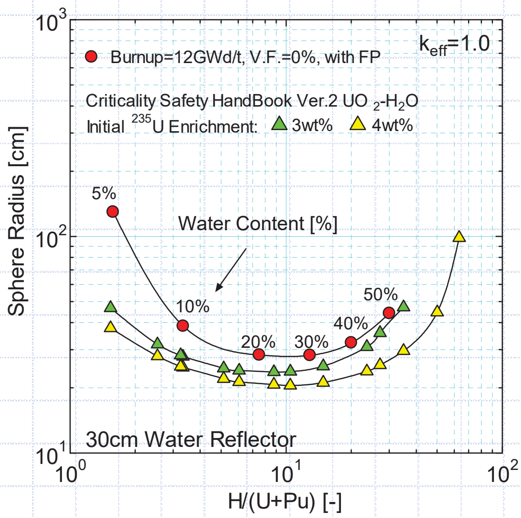


- Evaluation of basic parameters of fuel debris
  - Minimum criticality mass, radius
- OECD/NEA/WPNCS/EGBUC/ Phase-3B benchmark (BWR STEP-2 Fuel Burnup Calculation)
- JENDL-3.2
- BUC isotopes of EGBUC benchmarks.

Example of Minimum Criticality Size



Size of geometry to Reach Criticality



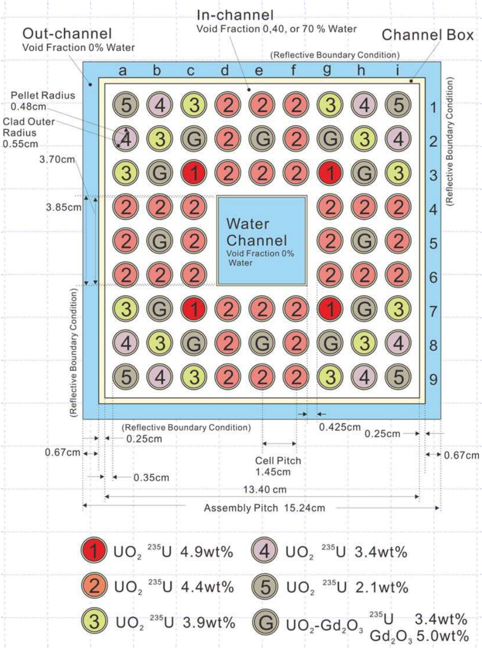
Comparison with Fresh Fuel Case

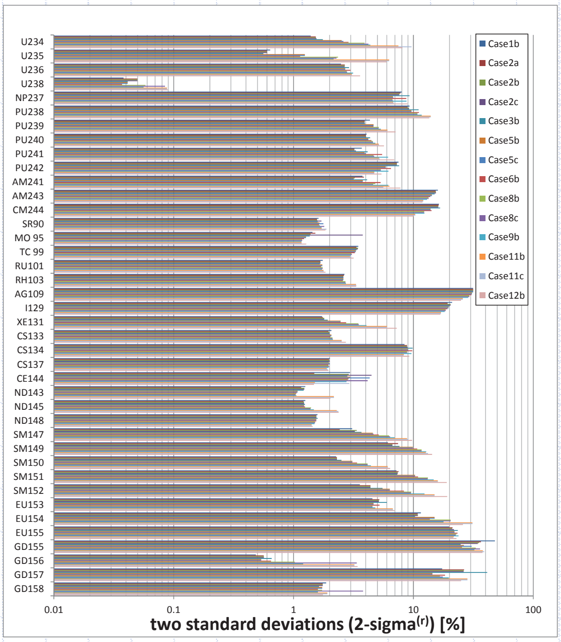
2012 AESJ Annual Meeting

Technical development in JAEA(2)

- International burnup calculation benchmark for BWR STEP-III fuel assembly under OECD/NEA/NSC/WPNCS/EGBUC framework.
- 35 sets of results from 16 institutes from 9 countries.
- To be published in 2015 from OECD/NEA

OECD/NEA/NSC/WPNCS/EGBUC Phase-IIIC Benchmark





Technical development in JAEA(3)

- Post Irradiation Examination(PIE) to measure assay data of used nuclear fuels of BWR and PWR
  - BWR – 1 sample
  - PWR – 9 samples
- TIMS, ICP-MS, New Chemical Separation, alpha-spectrometry
  - U, Np, Pu, Am, Cm
  - Tc, Rh, Ag, Cs, Nd, Sm, Eu, Gd

Post Irradiation Examination to measure isotopic composition

U	234	235	236	238			
Pu	238	239	240	241	242		
Nd	142	143	144	145	146	148	150
Np	237						
Am	241	243					
Cm	242	243	244	245			
★ Pm	147						
Sr	90						
(Zr)	93						
Ag	109						
Mo	95						
Tc	99						
Ru	101						
Rh	103						
Cs	133						
★ Sm	147	148	149	150	151	152	154
Eu	153	154	155				
Gd	154	155	156	157	158	160	

U, Pu, Nd → **TIMS** (Thermal ionization mass spectrometry)

Np, Am, Cm, Pm: Radiation and Mass Spectrometry

Sr, (Zr), Ag: Chemical Separation

Mo, Tc, Ru, Rh, Cs: Relatively easy to measure

Sr – Gd → **HR-ICP-MS** (High Resolution Inductively Coupled Plasma - Mass Spectrometry)

Enriched Spike → Isotope Dilution Method

Sm, Eu, Gd: Important Isotopes for Criticality Safety - chemical separation

Post Irradiation Examination to measure isotopic composition

Dissolved and Diluted Spent Fuel Sample

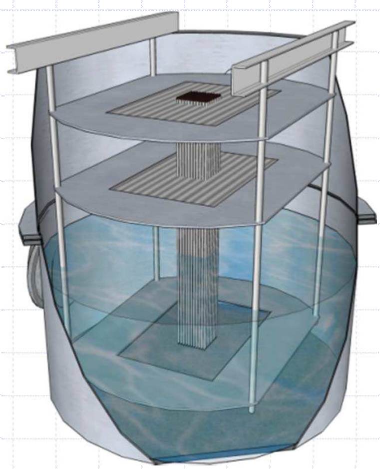
- 134,137Cs, 154,155Eu (γ-Spec)
- Am, Cm (α-Spec)
- Anion Exchange1 → U,Pu,Nd (TIMS)
- UTEVA Resin(remove U and Pu) → Mo,Tc,Ru,Rh, Cs
- Anion Exchange 2 → Ag
- Anion Exchange 3 → Gd,Eu,Sm,Pm,Nd
- HR-ICP-MS

In 2015

Except for 147Pm

Technical development in JAEA(4)

- Modification of STACY in NUCEF
  - (Homogeneous (solution) system to heterogeneous (fuel pin array))
  - Funded by Nuclear Regulation Agency of Japan
- Re-criticality of STACY is expected to be FY2018



The STACY modification has been funded by the Nuclear Regulation Authority (NRA) / the Secretariat of NRA of Japan.

Burnup Credit – is it feasible for Fukushima Fuel Debris?

To keep enough safety margin - again

- Kind of nuclear materials
  - What isotopes?
- Amount of nuclear materials
  - Nuclide density?

→ Conservative Isotopic Composition

- Consideration of physical and chemical status
  - Homogeneous/Heterogeneous(solution/array of rods)
  - uniform/un-uniform(concentration distribution)

Treatment of BUC in Japanese criticality safety handbook

- From initial publish in 1988, Japanese Criticality Safety Handbook (compiled by Nuclear Materials Regulation Division of Science and Technology Agency) **includes** the **idea** of BUC
  - *.. if the degree of burnup can be determined with high accuracy, then a criticality safety assessment may take the degree of burnup into account (1st version).*
  - *... however, when the evaluation of burnup is valid from the criticality safety evaluation viewpoint, the criticality safety evaluation may be conducted taking account of changes in nuclide composition resulting from burnup (2nd version).*
- Isotopic composition of uranium and plutonium (1st and 2nd versions)
  - Compilation of measured isotopic composition.
- Effect of burnup distribution should be considered separately (2nd version).
- FP nuclides that can be considered in criticality safety evaluation (2nd version).

Current Status of BUC in Japan

- BUC has already been introduced into the criticality safety evaluation at RRP of Japan;
  - SNF receiving pools (operated from 1999)
    - Non-destructive measurement to determine;
      - the burnup value of SNF and
      - remaining enrichment of uranium-235.
    - selection of a SNF storage rack of high capacity (BU< 2%) or not (2.5<BU<3.5%).
  - Dissolver
    - Measured burnup and initial enrichment
      - Addition of gadolinium nitrate solution in to dissolver for neutron absorber.

Status of BUC in Japan

	Wet Storage at Reactor	Interim Storage$	Transport	RRP	
				SNF receiving pool	Dissolver
PWR	N.A.	-	N.A.	(U,Pu)	(U,Pu)
BWR	G.C.	N.A.	N.A.	(U,Pu)	(U,Pu)

N.A.: Not Applicable, G.C.: Gd Credit is adopted instead of BUC, (U,Pu): U and Pu isotopes are considered in the credit, $Dry Storage system using metal casks. BWR fuel assemblies are considered.

BUC for Fuel Debris

- BUC or BUC-like method is considered to be introduced to relax the tough condition to control the criticality of Fukushima debris.
- It is a difficult task to take BUC for the fuel debris.
  - No method to evaluate the isotopic composition because of too many variety of composition (U, Pu, MA, FP)
  - Wide range of burnup for fuel debris (can we measure the burnup?)
  - Do all Fission Products exist in Fuel Debris (volatility)?
- Is it possible for us to introduce BUC in Fukushima Debris criticality safety control even though we have not yet adopted BUC in the intact fuel transport and SFP storage?

Treatment of FP should be different from intact fuel / reprocessing case.

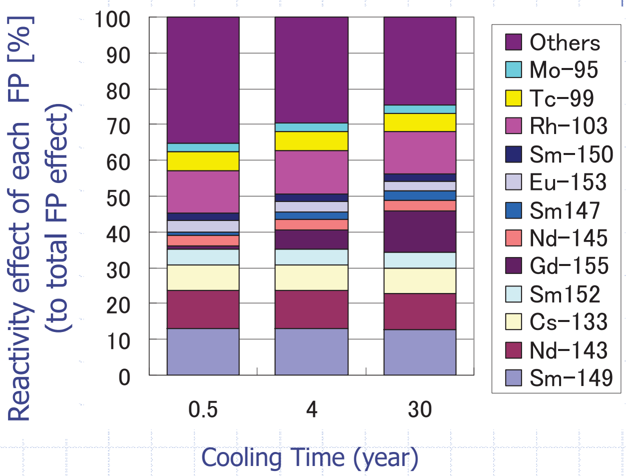
Selection of FP to be included into BUC in Japan

- Long half life or stable
- Non volatile
- Large reactivity effect

- *JAERI-Tech 2001-055* – more than 65% reactivity of total FP's contribution

^{149}Sm, ^{103}Rh*, ^{143}Nd, ^{133}Cs, ^{99}Tc*, ^{152}Sm, ^{155}Gd, ^{145}Nd, ^{147}Sm, ^{95}Mo*, ^{153}Eu, ^{150}Sm

*exclude after dissolution process



After SA, can we confirm the existence of FP with fissile?

Conclusion

Conclusion

- Criticality safety control is one of the key issues for decommissioning of the Fukushima site.
- We need clear strategy and consensus on this issue.
- IRID is the framework organizing R&D on Fukushima decommissioning.
- JAEA has been carrying out relevant research activities.
- Burnup Credit is one of the option for the criticality safety control of the Fuel Debris. But not easy way.