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Deconsolidation of HTR-10 Irradiated Fuels & Measurement of Burnup for SFE Storage

Xiaotong CHEN*, Bing LIU*, Taowei WANG,
chenxiaotong@tsinghua.edu.cn
bingliu@tsinghua.edu.cn
Institute of Nuclear and New Energy Technology, Tsinghua University, Beijing 100084, China

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- Dr. Xiaotong CHEN, Associate Professor
- Division of New Materials, Institute of Nuclear and New Energy Technology, Tsinghua University, Beijing, China
- Research fields
  - Performance evaluation of advanced fuel elements
  - Processing of HTR spent fuel element
  - Source term analysis of HTR irradiated fuels

OUTLINE

- Introduction
- Deconsolidation and Measurement
- Burnup uniformity in HTR-10 SFE
- Comparison of burnup calculation
- Conclusions

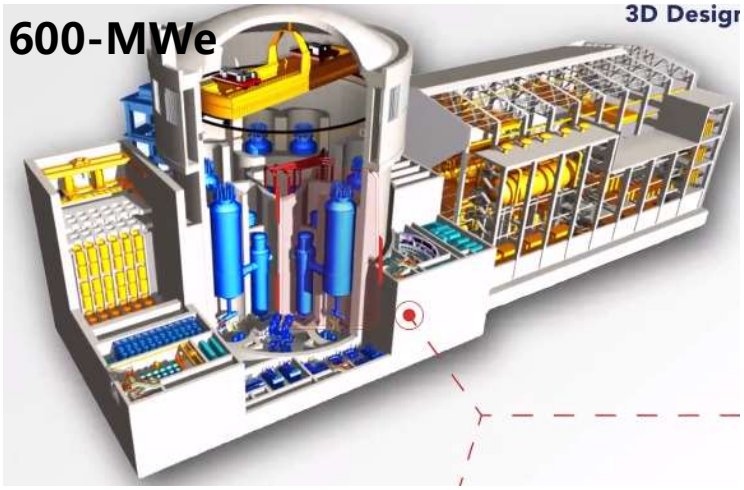
• Introduction

HTR spherical fuels (with TRISO particles)

	U-loading	Other indexes
HTR-10	5g/FE	Almost the same
HTR-PM	7g/FE	
HTR-PM600	7g/FE	

HTR-10 in INET of Tsinghua University, Beijing, China
1st criticality in 2000

HTR-PM demonstration plant in Shidao Bay Shandong Province, China
1st concrete poured on Dec 9, 2012
1st criticality on Sep 12, 2021



3D design of the HTR-PM600 nuclear power plant
Design completed in 2016

- The commercialization of HTGR in China highlights the storage challenge of SFEs.
- The burnup measurement of SFE is an important issue.

• Introduction

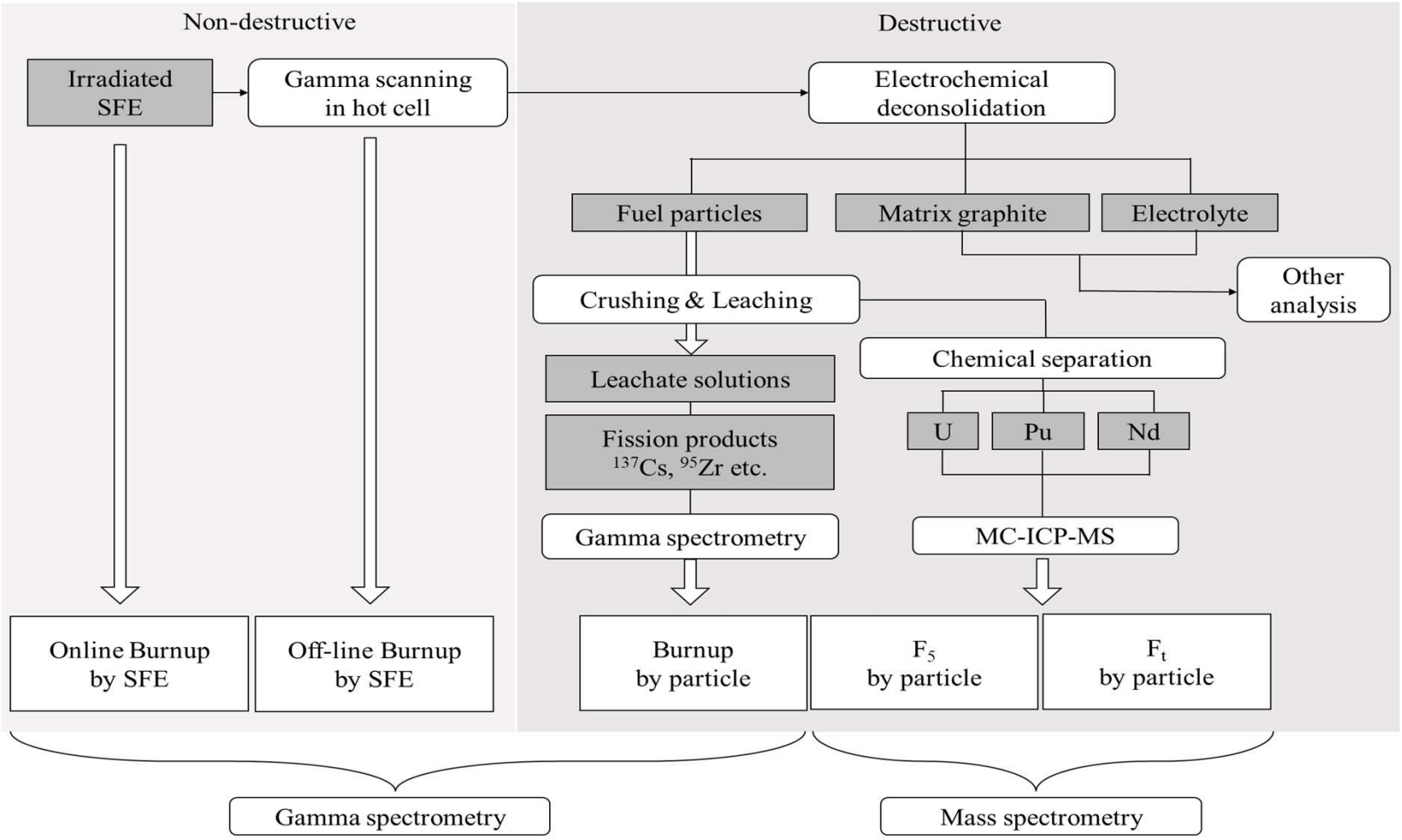
- This work aims for burnup measuring of SFEs. Three SFEs with low to medium burnup were selected and measured using destructive and non-destructive methods.
- Gamma and mass spectrometry were applied, and an electrochemical deconsolidation process was conducted to obtain TRISO fuel particles from specific regions.
- The uniformity of burnup in each SFE was also studied simultaneously by the mass spectrometry method.

HTR fuels before (left) and after (right) irradiation

INET HOTLAB (for PIE of HTR fuel pebbles)

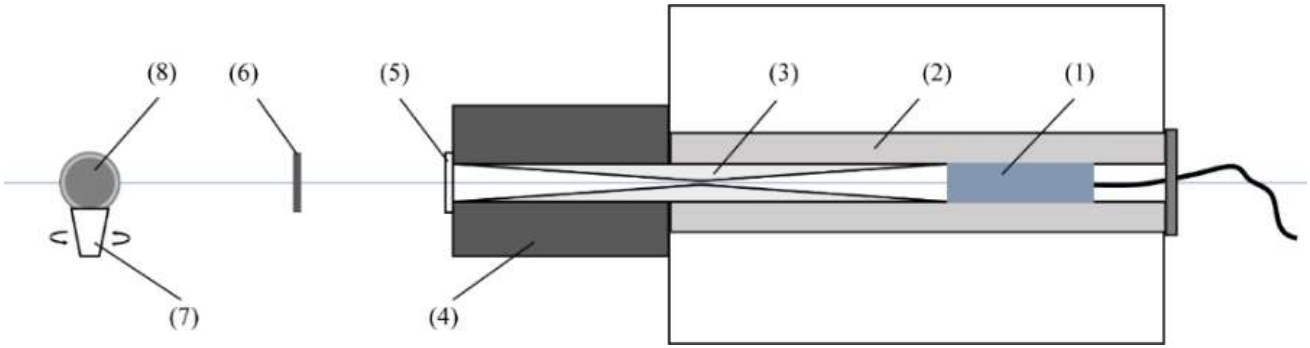
Deconsolidation & Measurement

Flow chart of the experimental process



Deconsolidation & Measurement

Gamma spectrometry method



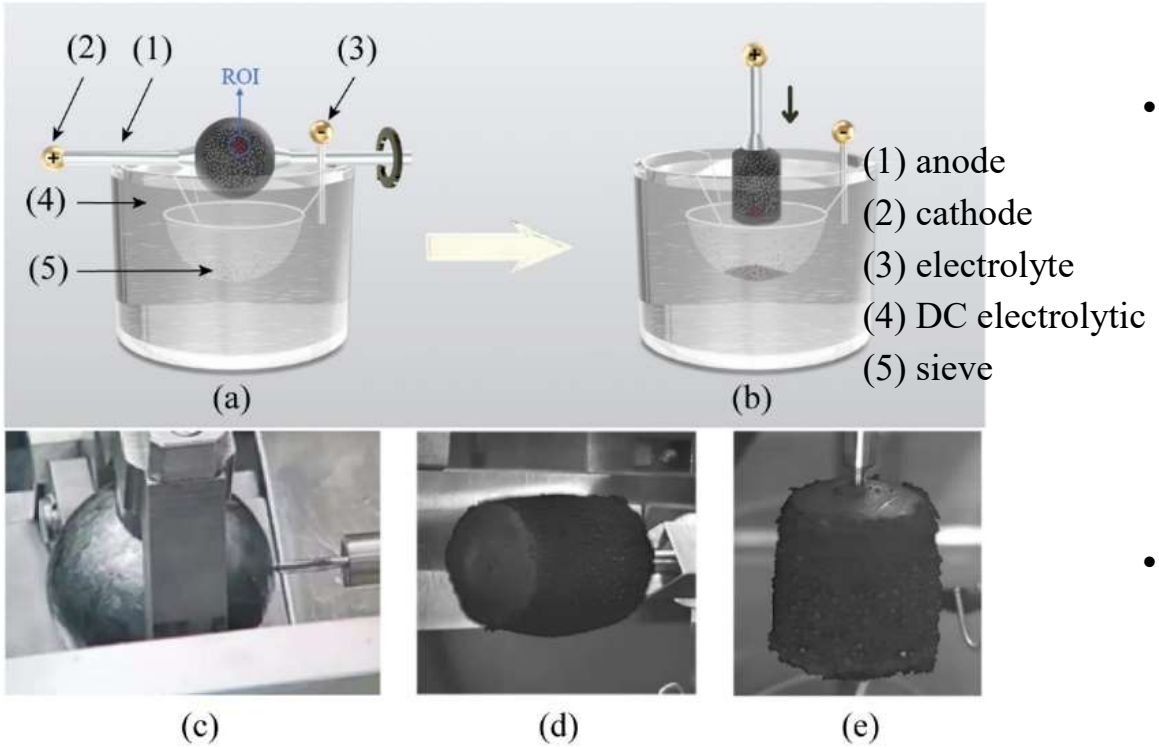
The burnup measurement system in the hot cell

(1) HPGe detector (2) collimator (3) penetrating sleeve
(4) shielding blocks (5) sealing window (6) shielding disks
(7) a rotating stage (8) SFE

- Gamma spectrum of three SFEs (P1, P2, P3) was first collected by BUMS before unloaded from HTR-10 and transferred to the hot cell, where provided a much lower radiation background and longer live time than at the reactor.
- Radioactivity of the caesium fission product was sophisticatedly measured.

Deconsolidation & Measurement

Deconsolidation and particles separation



The electrochemical deconsolidation process of an irradiated spherical fuel element (a, d) first stage deconsolidation (b, e) second stage deconsolidation (c) connecting rod assembling

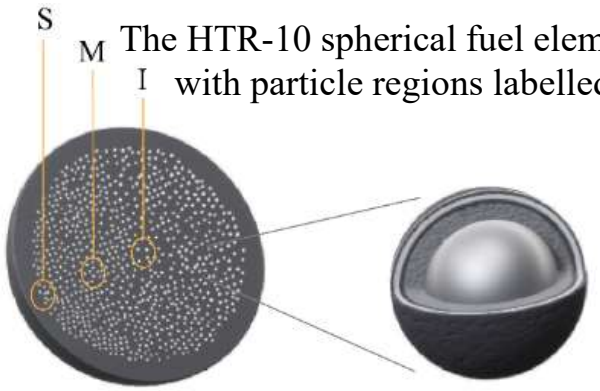
- The primary technical route for electrochemical deconsolidation of SFE is based on the anodic oxidation of strong acids.
- The mechanism is anion's transporting towards the anodic graphite matrix and intercalate into the graphite layer to form graphite intercalation compounds (GICs) under concentration and electrostatic potential gradient.
- These GICs are not stable and decompose naturally in the presence of water, loosing TRISO fuel particles and separating most graphite matrix powders attached.

Deconsolidation & Measurement

Leaching and Mass Spectrometry

- After the deconsolidation and separation, dozens of particles were randomly selected from different regions of the fuel zone and labelled as Surface, Middle and Inner.
- The selected particles were crushed to expose UO_2 kernels and transferred for the leaching process.
- Spectrometry (MC-ICP-MS, Neptune plus) was used to measure the content and isotope ratio of uranium, neodymium and plutonium nuclides.

Fuel element

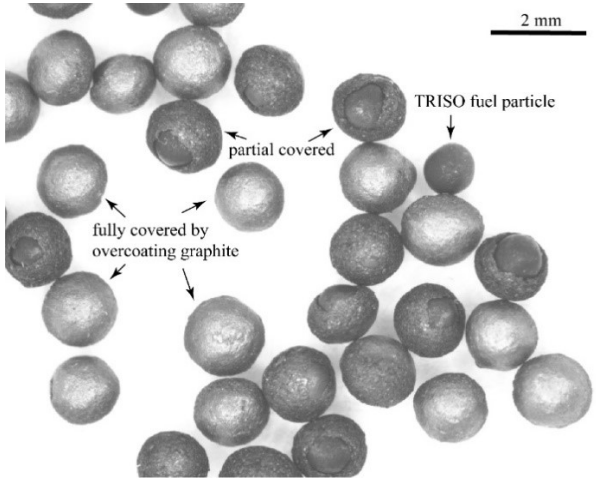


Fuel element cross-section

TRISO particle

The HTR-10 spherical fuel elements with particle regions labelled

TRISO fuel particles achieved after electrochemical deconsolidation



Burnup uniformity in SFE

$$F_5\ (\%) = N_8^0\left[\left(R_{5/8}^0 - R_{5/8}\right) - \left(R_{6/8} - R_{6/8}^0\right)\right]$$

N_8^0 : atom percentage of 238U in the pre−irradiated fuel
$R_{5/8}^0$: atom ratio of U-235 to U-238 in the pre-irradiated fuel
$R_{5/8}$: atom ratio of U−235 to U−238 in the irradiated fuel
$R_{6/8}^0$:atom ratio of U-236 to U-238 in the pre-irradiated fuel
$R_{6/8}$: atom ratio of U-236 to U-238 in the irradiated fuel

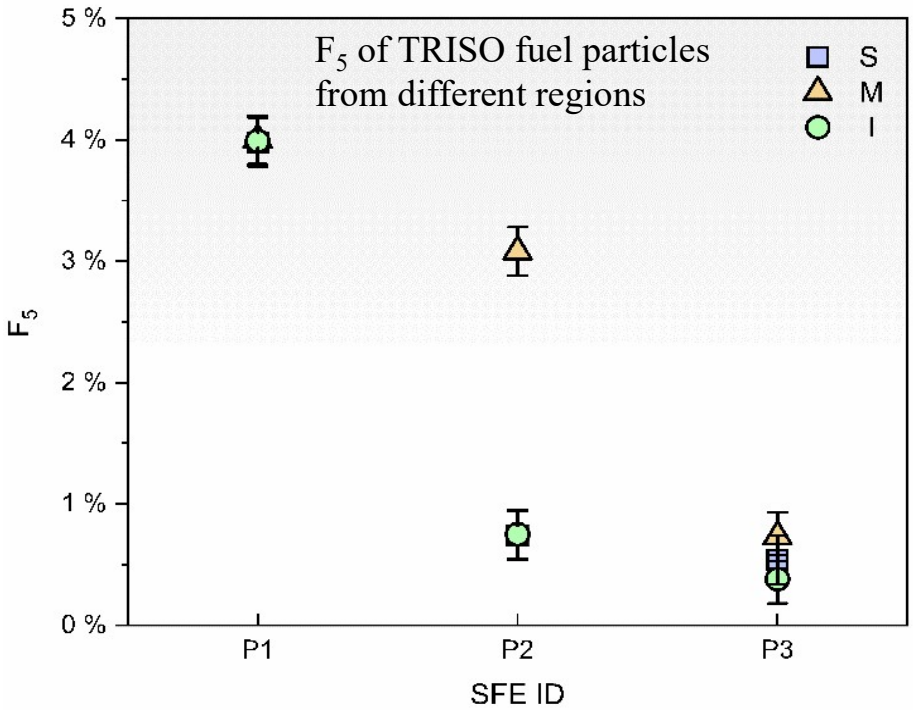


Table Atom ratio of uranium isotopes in irradiated fuel

Batch No.		$R_{5/8}$	$R_{6/8}$
P1	S	1.44E-01	1.29E-02
	M	1.44E-01	1.31E-02
	I	1.44E-01	1.30E-02
P2	S	1.91E-01	4.71E-03
	M	1.57E-01	1.04E-02
	I	1.91E-01	4.73E-03
P3	S	1.97E-01	1.52E-03
	M	1.94E-01	2.58E-03
	I	1.99E-01	1.51E-03

- After the deconsolidation and separation, dozens of particles were randomly selected from different regions of the fuel zone and labelled as Surface, Middle and Inner.
- The selected particles were crushed to expose UO_2 kernels and transferred for the leaching process.
- Spectrometry (MC-ICP-MS, Neptune plus) was used to measure the content and isotope ratio of uranium, neodymium and plutonium nuclides.

Burnup uniformity in SFE

Distribution of Cs-137

- The total activity of Cs-137 in each irradiated SFE was measured separately by non-destructive gamma spectrometry and destructive gamma spectrometry of particle leachate solutions and remnants.
- Table 3 lists the directly non-destructive to destructive derived activities ratio (N/D) of Cs-137.Differences caused by reasons of U-loading in one SFE, the deviation of UO_2 kernel diameter uranium contamination during fabrication and were calculated, and the ratio ranges among 0.92~1.09, which covers most values.

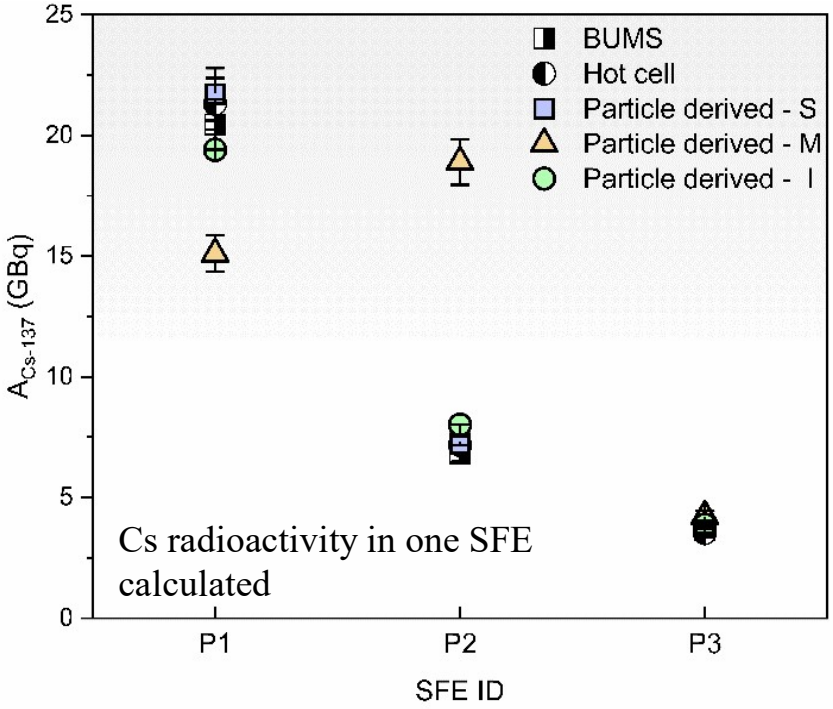


Table 3 Measured to derived activity ratio

Batch No.		N/D
P1	S	1.03
	M	0.72
	I	0.92
P2	S	1.05
	M	2.74
	I	1.11
P3	S	1.06
	M	1.12
	I	1.07

Comparison of burnup calculation

Caesium indicators

$$BU = \frac{A_{137}(t_{irr})}{\gamma_{137} N_{HM}} \left(1 - e^{(-\lambda_{137} t_{irr})}\right)^{-1}$$

$$BU = \frac{A_{137}}{\lambda_{137} \gamma_{137} N_{HM}}$$

BU:burnup in fissions per initial metal atoms (FIMA)

$BU = 1.7023 \times 10^{-12} A_{137}$

$A_{137}(t_{irr})$: radioactivity of Cs-137 at time t_{irr}

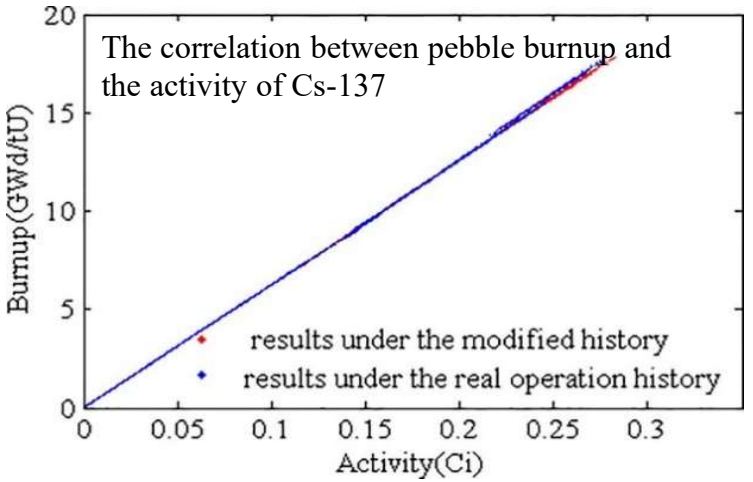
$A_{137-EOI}$: radioactivity of Cs−137 at the end of irradiation

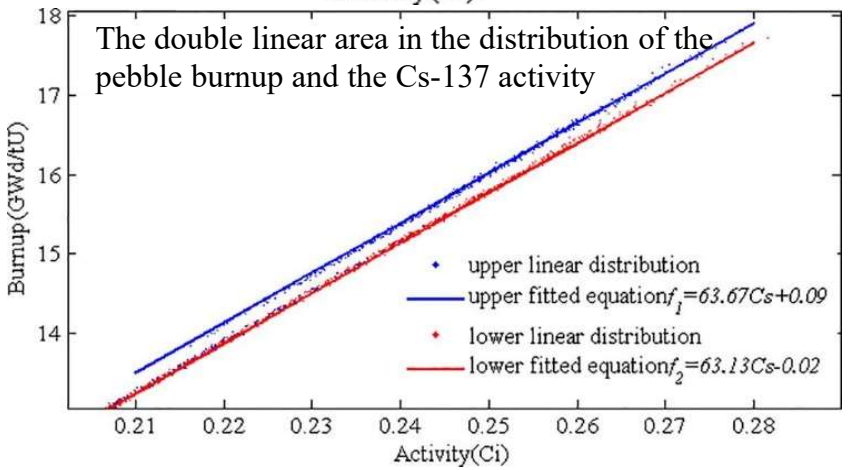
t_{irr}:irradiation time

λ_{137}:7.3021×10^{-1} s^{-1}, decay constant of Cs-137[36]

γ_{137}:0.0634, fission yield of Cs-137 in HTR-10[36,37]

N_{HM}:1.2689×10^{22}, initial number of heavy metal atoms[2]





Comparison of burnup calculation

Neodymium indicators

$$F_t = \frac{N/Y}{N/Y + N(U) + N(Pu)}$$

$$Y = \frac{1}{2}\left[\left(\frac{\sum_i N_i \sigma_i Y_i}{\sum_i N_i \sigma_i}\right)_0 + \left(\frac{\sum_i N_i \sigma_i Y_i}{\sum_i N_i \sigma_i}\right)_{End}\right]$$

N:number of Nd-148 or Nd-145 + Nd-146 atoms

$N(U)$:number of Nd-148 or Nd-145 + Nd-146 atoms

$N(Pu)$:number of Nd-148 or Nd-145 + Nd-146 atoms

Y: number of Nd−148 or Nd−145 + Nd−146 atoms

N_i:number of fissile isotopes

σ_i: fission cross section of fissile isotopes

Y_i:fission yield of the monitor isotope towards fissile isotopes

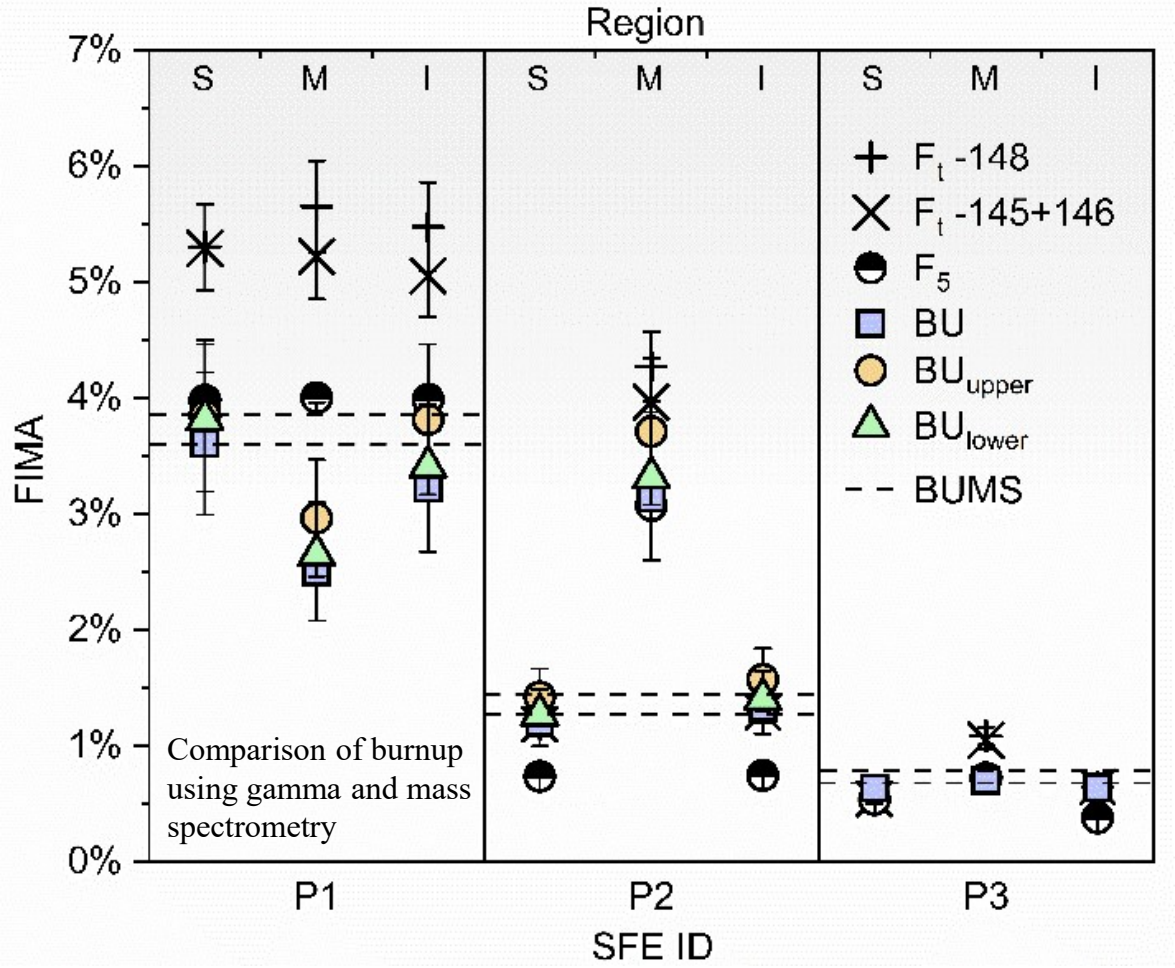
Neutron fission yield of Nd isotopes from U and Pu

Nuclides	Thermal neutron fission yield %		
	Nd-145	Nd-146	Nd-148
U-235	3.9439	2.9866	1.71
Pu-239	3.0357	2.4963	1.85

Fission cross section of U and Pu

Nuclides	σ (n,f)
U-235	112.37
Pu-239	356.31

Comparison of burnup calculation



- The burnup values derived from radiometric and chemical methods were compared.
- The reliability of using Cs-137 as an indicator for HTR-10 online burnup monitoring is confirmed.
- Factors of inadequate recovery of uranium from irradiated particles, transmutation of actinide, anddecay of Cs-137 may contribute to the disagreement in P1.

Conclusions

- Three irradiated spherical fuel elements from HTR-10 were electrochemically deconsolidated.
- The main FPs like U-235, U-238, Cs-137, Nd-148 were measured by the radiometric and mass spectrometric methods.
- The fractional U-235 burnup was used to determine the burnup distribution, which proved that in each SFE, the burnup is relatively uniform.
- The measured burnup using gamma spectrometry by Cs-137 compared well with the one using mass spectrometry by uranium nuclides.

THANKS FOR YOUR ATTENTION!