53rd Annual Meeting HOTLAB2016

Examination of analytical method of rare earth elements in used nuclear fuel

Back-**E**nd fuel **C**ycle **K**ey elements research facilit**Y**

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Contents

1. Background (1/2)

- For criticality safety evaluation,
  - The burnup value is important and indispensable.
  - Burnup calculation codes are being continuously improved or developed.
  - It is required to evaluate the amounts of fission products in used nuclear fuel.

- Some isotopes of rare earth elements such as Gd, Eu and Sm have large neutron absorption cross sections and high fission yield.
- The amounts of these isotopes are computed by burnup calculation codes for criticality safety evaluation.

Some isotopes of rare earth elements have same mass number.

Objective

To separate rare earth elements prior to the measurement by mass spectrometry.

1. Background (2/2)

- For the evaluation of burnup,
  - JAEA had developed an analytical method to measure amounts of U, Pu and Nd.

"Conventional method"

We examine to measure the rare earth elements such as Gd, Eu and Sm **by improving the conventional method**.

- To obtain isotopic composition data of used nuclear fuel including the rare earth elements in addition to U, Pu and Nd without complicated process added.
- To obtain these data from one sample.

2. Improvement of the analytical method

1. Background

2. Improvement of the analytical method
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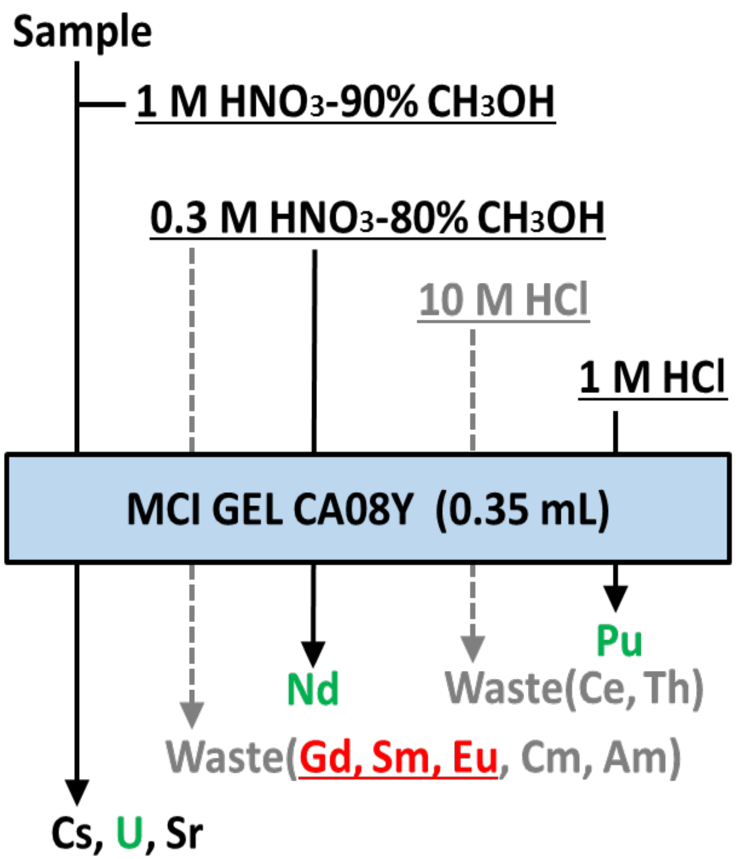
4. Conclusion

2.1 Conventional separation method

- Quartz column; inner diameter of 3mm
- Anion exchange resin; MCI GEL CA08Y, volume of 0.35 mL
- Temperature; 26˚C during elution with thermostatic bath

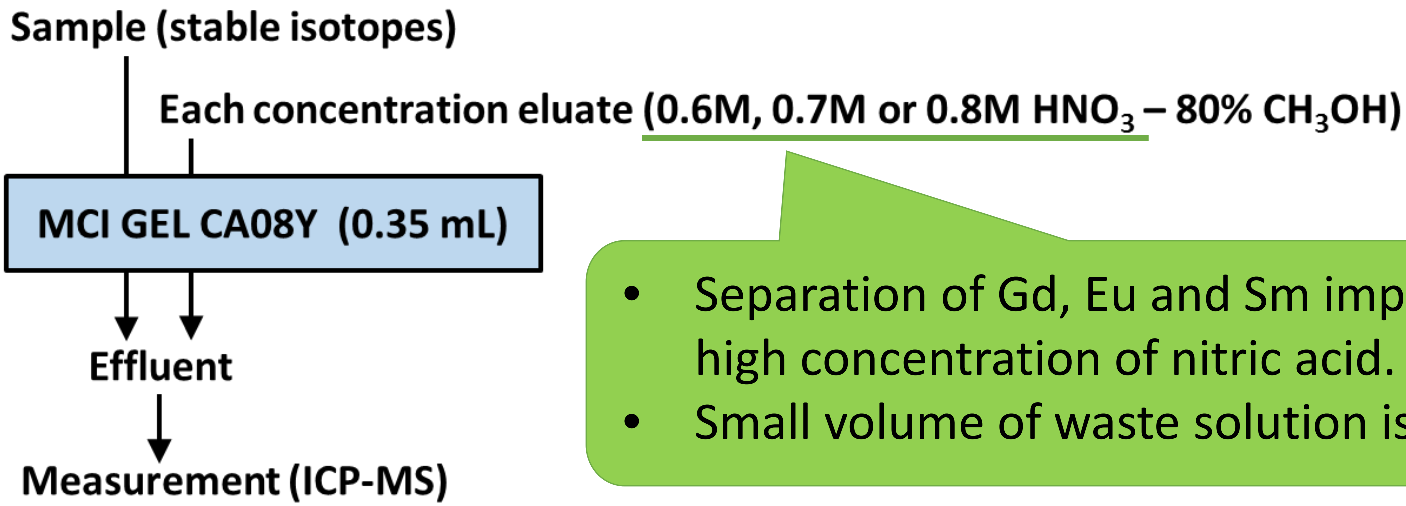


➢ Conventional method

- Suitable method to separate **U, Pu** and **Nd**.
- Rare earth elements such as **Gd, Eu** and **Sm** except Nd are washed out to waste portion.

We examined to separate these elements in waste portion by adding another eluate of different chemical composition.

2.2 Experimental condition of separation



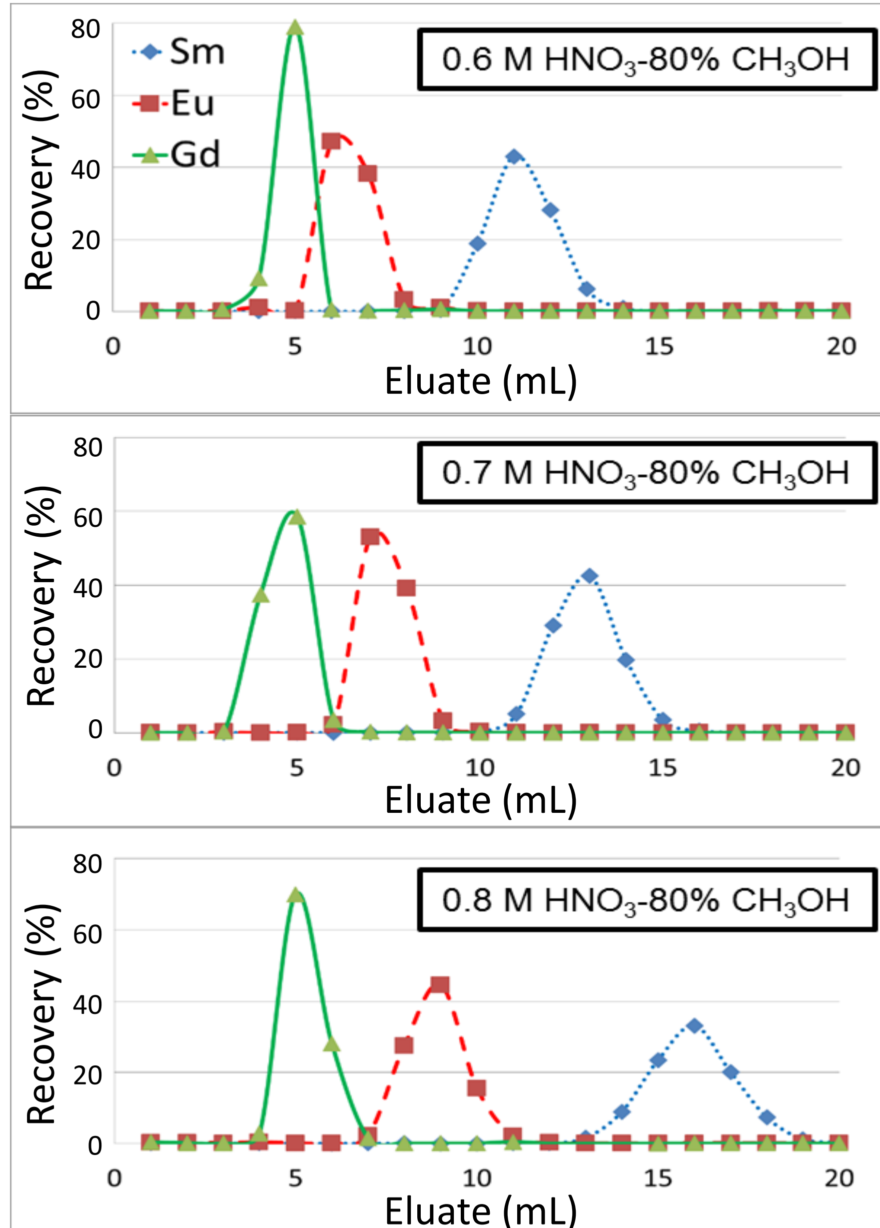
- Separation of Gd, Eu and Sm improves with high concentration of nitric acid.
- Small volume of waste solution is also required.

Amounts of samples	^{149}Sm : about 200 pg ^{151}Eu : about 150 pg ^{155}Gd : about 350 pg
Eluate concentrations	1) 0.6 M HNO_3-80% CH_3OH 2) 0.7 M HNO_3-80% CH_3OH 3) 0.8 M HNO_3-80% CH_3OH
Feed sample condition	HNO_3 0.4 mL + CH_3OH 4 mL
Temperature	26˚C
Analytical equipment	**I**nductively **C**oupled **P**lasma **M**ass **S**pectrometry (ICP-MS)

2.3 Result

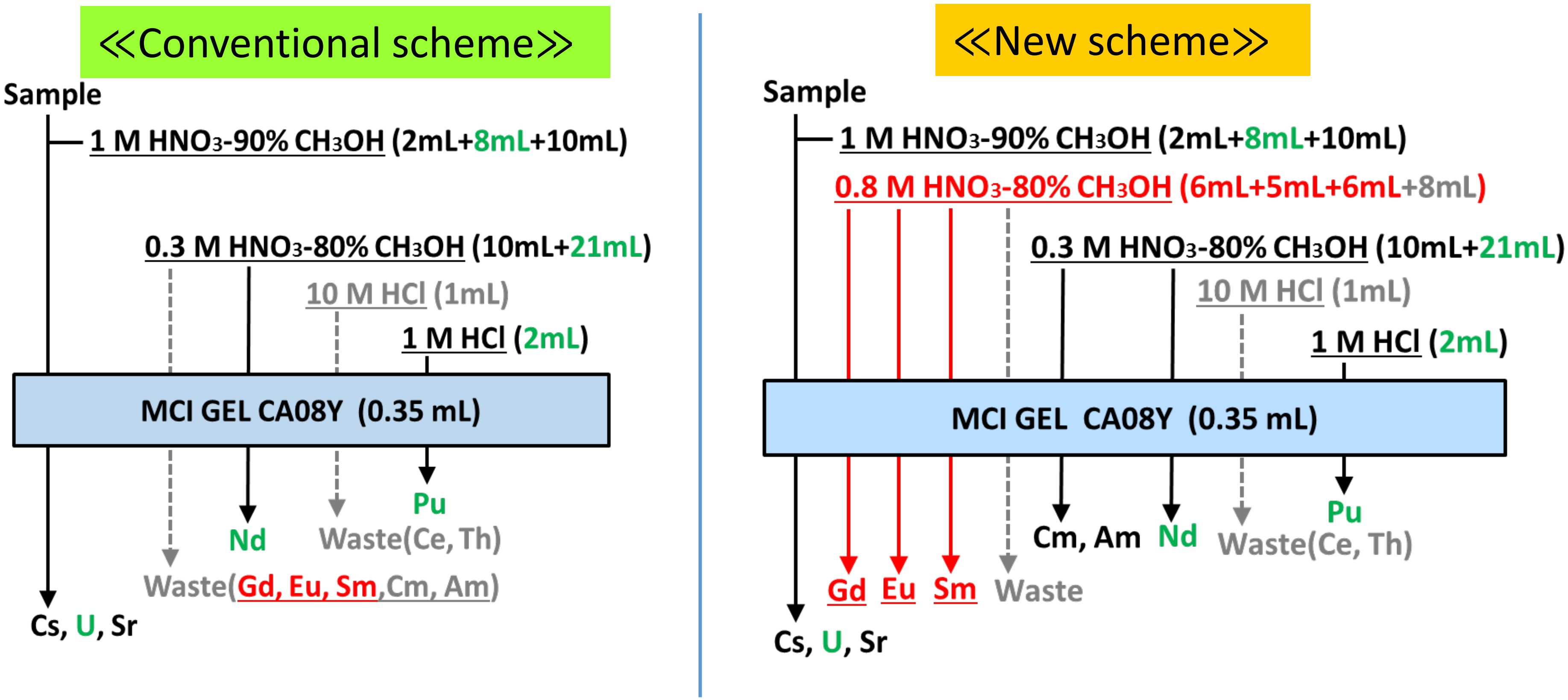
- Separation of Gd, Eu and Sm is enough with 0.8 M HNO_3 - 80% CH_3OH.
- Total volume of eluate is acceptable all cases.

We selected <u>0.8 M HNO_3 - 80% CH_3OH</u> among three different concentrations for the separation of Gd, Eu and Sm.



３. Evaluation of the improved analytical method

3.1 New separation scheme



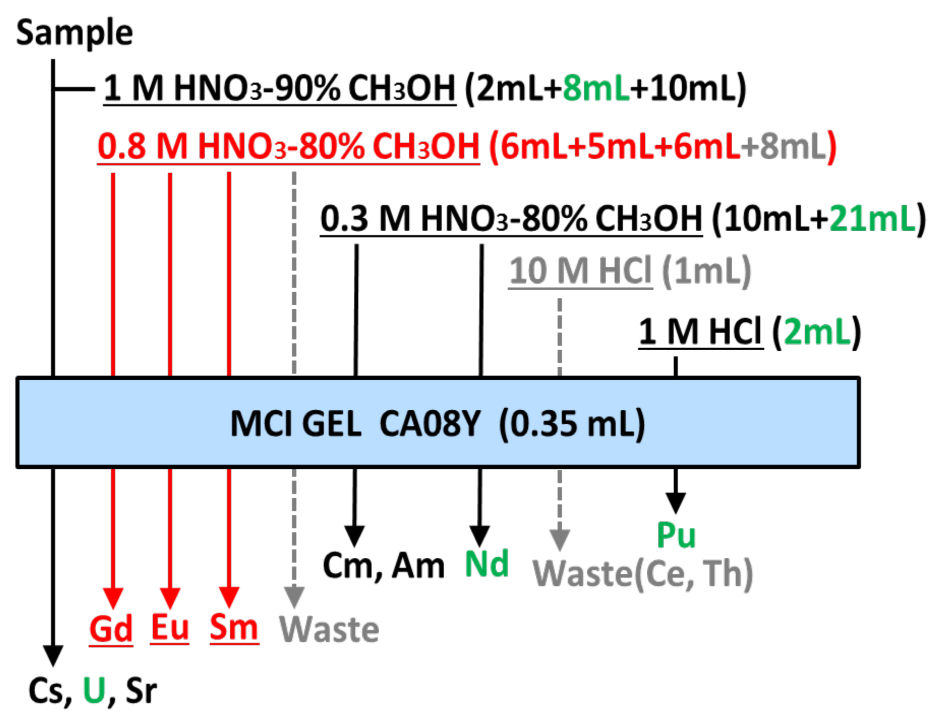
The separation process of Gd, Eu and Sm with 0.8M HNO_3-80% CH_3OH is added between the elutions of U and Nd.

We confirm the influence of the added process on elution of Nd and Pu.

3.2 Confirmation of new scheme

- U, Pu and Nd were used in order to confirm the influence of the newly added process.
- Amounts of these elements are about the same with the amounts included in the used nuclear fuel samples.

Amounts of samples	^{233}U : about 2 μg ^{242}Pu : about 50 ng ^{150}Nd : about 20 ng ^{149}Sm : about 200 pg ^{151}Eu : about 150 pg ^{155}Gd : about 350 pg
Feed sample condition	HNO_3 0.4 mL + CH_3OH 4 mL
Temperature	26˚C
Analytical equipment	ICP-MS



3.3 Result (1/2)

Fractions	Recovery (%)							
	The conventional method				The new method			
	^{233}U	Gd, Eu, Sm	^{150}Nd	^{242}Pu	^{233}U	Gd, Eu, Sm	^{150}Nd	^{242}Pu
U fraction	71.4	< 1	< 1	< 1	74.1	< 1	< 1	< 1
Nd fraction	< 1	< 1	97.7	29.8	< 1	< 1	96.4	36.6
Pu fraction	< 1	< 1	< 1	62.9	< 1	< 1	< 1	52.9

- Elements which interfere with the measurement of the target did not elute in each fraction.
- Recovery rates of U, Pu and Nd are almost same between the conventional method and the new method.

➡ The newly added separation process did not interfere with separation and measurement of U, Pu and Nd.

3.3 Result (2/2)

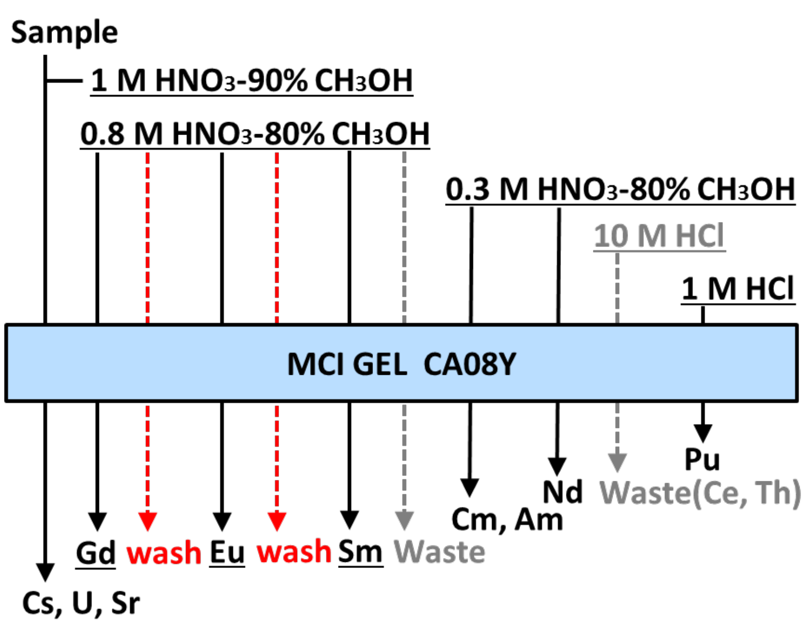
Fractions	Recovery (%)		
	^{155}Gd	^{151}Eu	^{149}Sm
Gd fraction	93.0	25.1	< 1
Eu fraction	4.4	77.9	1.3
Sm fraction	< 1	< 1	91.4

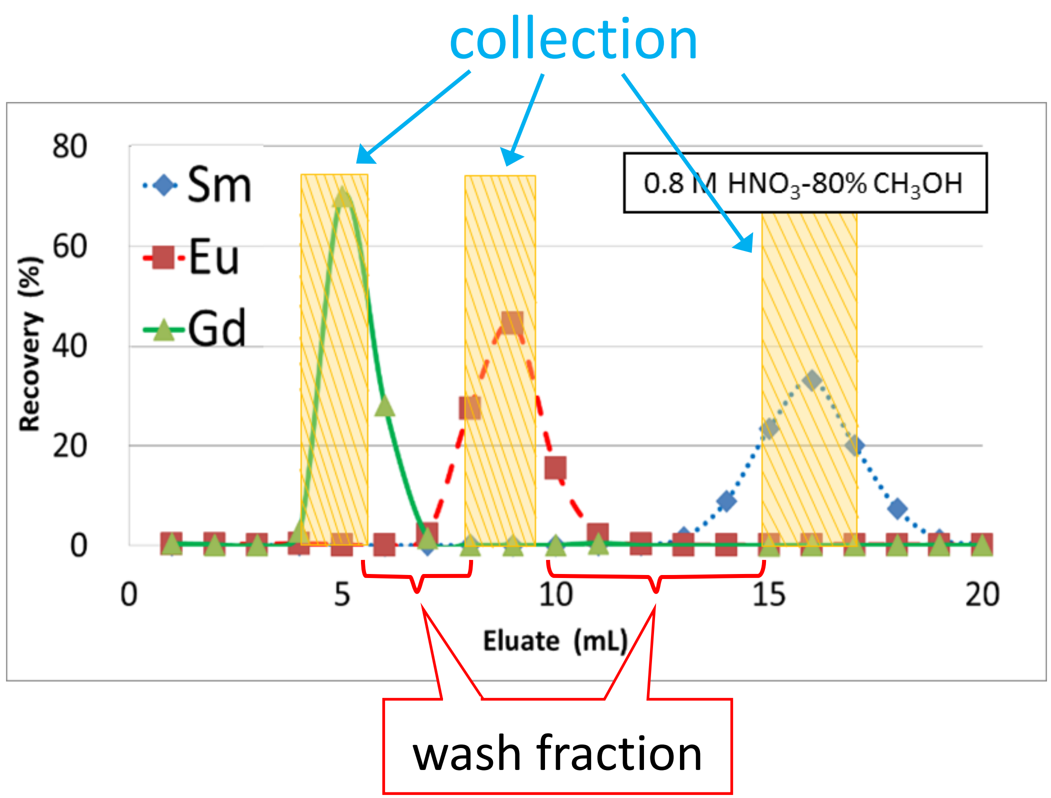
- The new method succeeded to separate most of Gd, Eu and Sm.
- Eu eluted in Gd fraction, and Sm and Gd eluted in Eu fraction.

  \- High recovery is not necessary, but high purity is required for the measurement of each element by IDMS method.

《Future work》

- Sampling near the boundary of each element should be avoided, especially before and after Eu fraction.
- It is required to collect effluent <u>near the peaks of each element</u> for the measurement with highly pure samples.





4. Conclusion

- We obtained prospect for separation of Gd, Eu and Sm by the new method.
- The added separation process of rare earth elements did not interfere with separation of U, Pu and Nd.

- The new method made it possible to obtain isotopic composition data of used nuclear fuel including the rare earth elements.
- The data obtained by the new method will be used for:
  - Validation and improvement of burnup calculation codes.
  - Burnup credit calculation of used nuclear fuel.