# 8–15 Toward the Rapid Analysis of Plutonium Isotopes

## —Analytical Technique Without Pretreatments—

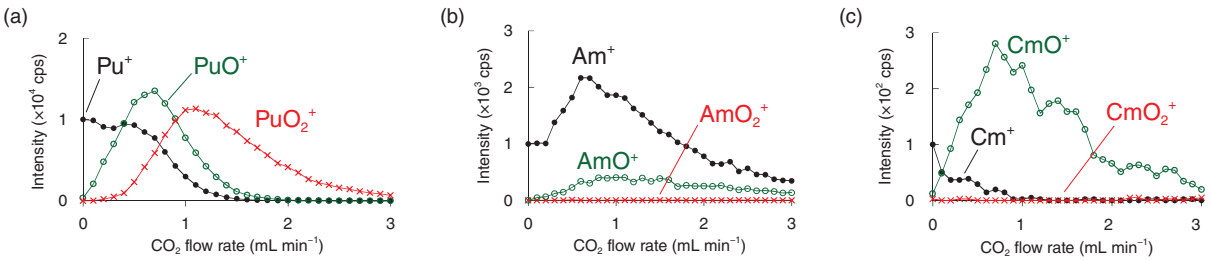


**Fig.1 Reactivity of actinides with $CO_2$ gas**
Pu is oxidized to a dioxide form by $CO_2$, though Am and Cm form monoxides. This implies that isotopes with the same mass number can be separated by taking advantage of the different progressions of oxidation.

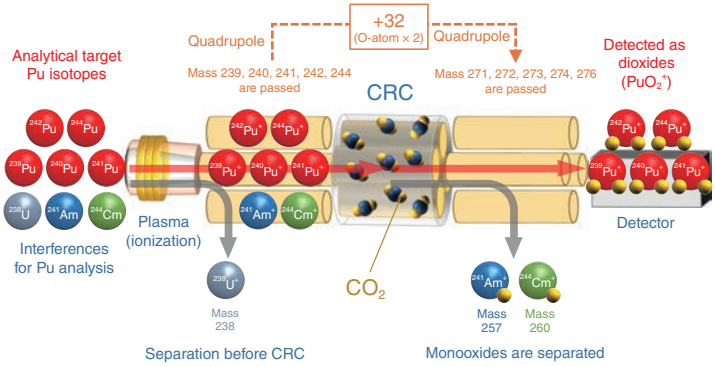


**Fig.2 Concept of simultaneous determination of Pu isotopes**
The quadrupole present prior to the collision reaction cell (CRC) removes $^{238}U$ to avoid the creation of $^{238}UO_2$ hydride at the CRC. Pu, Am, and Cm react with $CO_2$ at the CRC. Pu passes through the quadrupole of the back end of the dynamic reaction cell (DRC) as a dioxide, while Am and Cm are removed as monoxides. By this technique, Pu isotopes ($^{239}Pu$, $^{240}Pu$, $^{241}Pu$, $^{242}Pu$, and $^{244}Pu$) can be simultaneously detected via one-shot data acquisition.

Pu isotopes are important radionuclides to manage radiation exposure control and radioactive waste at the TEPCO's Fukushima Daiichi Nuclear Power Station. Measurement of Pu isotopes ($^{239}Pu$, $^{240}Pu$, $^{241}Pu$, $^{242}Pu$, and $^{244}Pu$) is necessary to obtain information on distribution and contamination levels. However, the conventional method using alpha-ray spectrometry requires a considerably long analytical time and high levels of efforts and techniques for separating radionuclides with energy similar to that of the alpha rays emitted by Pu.

Inductively coupled plasma–mass spectrometry (ICP-MS) allows measuring multiple radionuclides simultaneously and rapidly via mass spectrometric discrimination without any separation steps requiring handling by humans. However, the quantification is affected by co-existing isobaric interferences. $^{239}Pu$ and $^{240}Pu$ overlap with $^{238}U$ hydride ($^{238}UH^+$ and $^{238}UH_2^+$) and $^{241}Pu$ and $^{244}Pu$ with $^{241}Am$ and $^{244}Cm$, respectively. In this study, we developed a discrimination method to detect multiple Pu isotopes simultaneously in a single data acquisition using ICP-tandem mass spectrometry (ICP-MS/MS), in which the instrument is equipped with a collision reaction cell (CRC) between two quadrupoles.

First, we introduced several oxidizing gases individually ($O_2$, $CO_2$, and NO) into the CRC and investigated the oxidation behaviors of Pu, Am, and Cm. Pu was converted to dioxides ($PuO_2^+$) and Am and Cm were converted to monoxides ($AmO^+$ and $CmO^+$) by $CO_2$ (Fig.1), making mass separation of $^{241}Pu$ and $^{244}Pu$ from $^{241}Am$ and $^{244}Cm$ possible. The $^{238}UO_2^+$ ion generated from $^{238}U^+$ by oxidation in the CRC creates hydrides ($^{238}UO_2H^+$ and $^{238}UO_2H_2^+$) that interfere with $^{239}PuO_2^+$ and $^{240}PuO_2^+$. Therefore, $^{238}U^+$ was removed at the quadrupole before CRC to suppress the formation of $^{238}UO_2$ hydride. The combined separation of isobaric interferences ($^{241}Am^+$, $^{244}Cm^+$, and $^{238}U^+$) achieved remarkably low background counts for the mass of $^{239}Pu$, $^{240}Pu$, $^{241}Pu$, and $^{244}Pu$ (as $PuO_2^+$). From these results, it is clear that Pu isotopes ($^{239}Pu$, $^{240}Pu$, $^{241}Pu$, $^{242}Pu$, and $^{244}Pu$) can be measured simultaneously in one-shot data acquisition (Fig.2). Note that $^{238}Pu$ is out of scope of this determination because it is removed along with $^{238}U$.

This method discriminates target Pu isotopes and their isobaric interferences without any human-handling separation process. Moreover, the simultaneous determination of Pu isotopes will lead to the development of a high-throughput analysis and contribute to the decommissioning of nuclear power stations.

(Makoto Matsueda)

**Reference**

Matsueda, M. et al., Using $CO_2$ Reactions to Achieve Mass-Spectrometric Discrimination in Simultaneous Plutonium-Isotope Speciation with Inductively Coupled Plasma-Tandem Mass Spectrometry, Chemistry Letters, vol.51, no.7, 2022, p.678–682.