4–8 Quantitation of the Difficult-to-Measure Nuclide Palladium-107

— Measurement of Palladium Obtained by Laser Irradiation of Spent Nuclear Fuel —

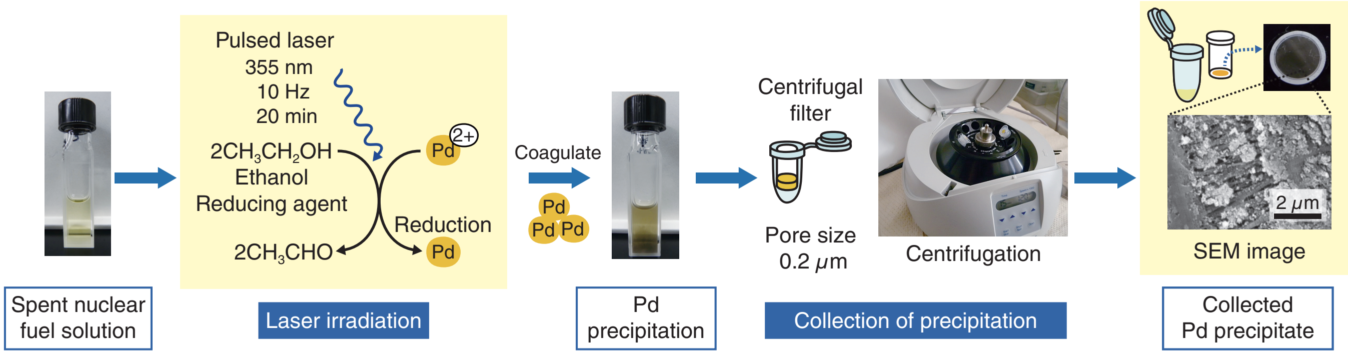

Fig.4-17 Separation mechanism of high-purity Pd by laser irradiation and its operating procedure
Laser irradiation induces reduction of a Pd ion, changing it from a divalent cation (Pd^{2+}) to a non-charged form. The reduced Pd atoms coagulate with each other to form a precipitate, allowing Pd to be easily recovered. This technique enables operators to maintain a safe distance from highly radioactive samples, leading to minimum radiation exposure.

Table 4-3 Contamination rates of major constituents of spent nuclear fuel in Pd precipitate
Almost no elements that initially existed in the spent nuclear-fuel sample were found in the Pd precipitate prepared by laser irradiation. Accurate quantitation of ^{107}Pd can only be achieved with such high-purity Pd.

Element	Sr	Cs	Ba	Ru	Rh	U	Np	Pu	Am
Contamination (%)*	<0.001	<0.001	0.001	<0.001	<0.001	0.004	<0.001	<0.001	<0.001

*Contamination (%) = (Weight of element found in Pd precipitate) / (Weight of Pd precipitate) × 100

Palladium-107 (^{107}Pd), which is one of the radionuclides generated by nuclear fission of uranium, is found in spent nuclear fuel. Due to its extremely long half-life of about 6.5×10^6 years, ^{107}Pd can be potentially harmful to human body over a long period of time. The long-term risks involved in ^{107}Pd radiation are basically estimated based on the ^{107}Pd content in spent nuclear fuel. However, predicted values provided through theoretical calculation have been used instead of the measured content of ^{107}Pd because of the lack of published information concerning ^{107}Pd quantitation.

Accurate measurement of ^{107}Pd can only be achieved by a separation technique which allows recovery of high-purity Pd from spent nuclear fuel. Chromatographic separation using adsorbent-packed columns, which is the most commonly applied technique, is inevitably associated with numerous operational steps, making it hard to complete the separation in a short time. Additionally, continuous work with highly radioactive samples, such as a spent nuclear-fuel solution, causes undesirable radiation exposure to workers.

The separation technique developed in this study enables selective recovery of Pd by a simple operation, leading to the world's first successful quantitation of ^{107}Pd in spent nuclear fuel. Precipitation of Pd is induced by irradiation by a pulsed laser of wavelength 355 nm (Fig.4-17). The precipitation reaction is completed in 20 minutes in a closed vessel and operators can keep their distance from a sample during irradiation. This can help minimize the contamination of laboratory facilities and radiation exposure to workers during irradiation.

The resulting Pd precipitate was dissolved with aqua regia (a mixture of nitric acid and hydrochloric acids) and measured by inductively coupled plasma-mass spectrometry. Almost none of the elements initially present were detected in the precipitate (Table 4-3) and Pd was recovered with a purity above 99.9%. With the perfect removal of all interfering components, ^{107}Pd in spent nuclear fuel was accurately measured.

Application of the developed technique to other samples, for example, high-level radioactive waste (HLW), is expected. We are trying to expand the versatility of our technique for application to HLW, which has a complicated chemical composition. We will make continuous efforts to establish highly reliable safety assessment of HLW disposal by offering accurate measurements of ^{107}Pd.

Reference

Asai, S. et al., Determination of ^{107}Pd in Pd Recovered by Laser-Induced Photoreduction with Inductively Coupled Plasma Mass Spectrometry, Analytical Chemistry, vol.88, issue 24, 2016, p.12227-12233.