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Novel Isotope Analysis Method Using a Supersonic Plasma Jet Combined with Diode Laser Absorption Spectroscopy

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Radioactive isotopes in radioactive waste must be quantified for environmental impact assessment and radiological protection of workers, which depend on their half-life, radiation energy, and radiation quality. In fact, radioactive isotopes (*e.g.* ^{90}Sr, ^{134}Cs, and ^{137}Cs) were released into the environment as a result of the accident at the Fukushima Daiichi Nuclear Power Plants in 2011, and a large amount of radioactive waste was generated.

Isotope analysis has been conventionally performed via inductively coupled plasma mass spectrometry (ICP−MS). Nevertheless, a commercial ICP−MS apparatus does not have sufficient resolution to avoid interference of isobars, and in the case of ICP−MS, complicated chemical separation, which typically takes 2−4 weeks, is necessary to prevent isobaric interference.

Furthermore, laser spectroscopy techniques using laser ablation exhibit high wavelength selectivity and can be employed for the rapid identification of radioactive elements without the need for chemical separation. A tunable diode laser with a narrow line width can be used to distinguish isotope shifts based on the mass number. Notably, laser ablation absorption spectroscopy (LAAS) has been developed for the direct isotope analysis of solid samples. Several recent studies describe the plasma plume dynamics and optimization of the LAAS conditions for direct isotope analysis. However, upon irradiation of the surface with an intense laser beam to atomize a solid sample, the translational temperatures of molecules and atoms emitted into a plasma plume reach 5000−10,000 K. Thus, for instance, the isotope shift of ^{88}Sr−^{90}Sr, 206.2 MHz, cannot be distinguished via LAAS due to an increase in the Doppler broadening under such high-temperature conditions. Considering natural broadening, the residual of the Doppler broadening must be suppressed to less than 180 MHz in the half width at half-maximum. This is equivalent to approximately 60 K in the translational temperature.

Thus, we have developed a novel isotope analysis method based on diode laser absorption spectroscopy (DLAS) using a supersonic wind tunnel to atomize samples and distinguish elements and isotopes directly without chemical separation.[1–3] The method is composed of two sections, namely, a high-temperature section for atomization of samples and a low-temperature section for spectroscopic measurements in a supersonic plasma jet to account for adiabatic expansion. The temperature reduction downstream of the supersonic nozzle due to adiabatic process provides a high wavelength resolution suppressing the Doppler broadening of each isotope for the detection of slight wavelength shifts between isotopes.

In our previous studies, we described the plasma properties in the two sections by using argon gas as the operating gas and demonstrate the analysis method by using xenon stable isotopes.[1] Next, stable Xe isotopes in the gaseous phase and stable Sr isotopes in the powder phase were investigated. Notably, dramatic reductions in the temperature of the Xe and Sr atoms to approximately 180 K and 85 K were achieved, and their corresponding resolutions are comparable with the slight isotope shifts due to the differences in mass number.[2,3] Moreover, based on the measured atomic number density and flow velocity, we found that the atomization efficiency of the Sr powder was approximately 10.4%.[3] These findings could pave the way for the development of a direct and rapid analysis method.

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References

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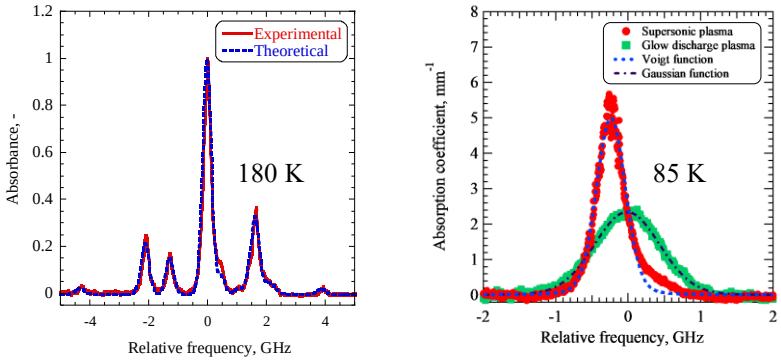

Figure 1. Typical absorption spectra of Xe stable isotopes (left) and Sr stable isotopes (right). The translational temperatures were calculated from the Doppler widths.