液体電極プラズマ発光分光法による再処理工程試料中の金属元素の分析技術開発 （2）高放射性廃液中のセシウム、テクネチウムの定量

Development of analytical methods for metal elements in reprocessing solution by optical emission spectrometry based on liquid electrode plasma

(2) Determination of cesium and technetium in highly active liquid waste

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A simple and rapid method for determination of Cs and Tc in highly radioactive liquid waste (HALW) using liquid electrode plasma (LEP) has been developed. Effects of pulsed voltage sequence and nitric acid concentration on emissions of these elements are investigated. The method is applied to the quantitative analysis of Cs and Tc in HALW samples at Tokai reprocessing plant.

Keywords: Liquid electrode plasma, cesium, technetium, highly radioactive waste, elemental analysis.

1. Introduction The introduction to our LEP system was presented in the first part. The second section focuses on the employment of the novel technique for determination of Cs and Tc, two important fission products in HALW samples.

2. Experiment The measurement method was systematically developed. The basic condition for generating plasma was optimized including voltage, pulse sequence (on-time, off-time and number of pulses). The effect of nitric acid concentration was investigated. The developed method was validated using a simulated sample in comparison with reference method, and finally applied for real HALW samples obtained from Tokai reprocessing plant.

3. Results and discussion We found the best condition for the quantification of Cs. There is no significant interference found at emission line of 852.11 nm using LEP. The voltage, on-time, off-time and number of pulses were 800 V, 2 ms, 1 ms and 30 pulses, respectively. Nitric acid with a concentration of 0.4 M offered the best reproducibility for Cs emission. Limit of detection (LOD) and limit of quantification (LOQ) were determined to be 0.005 mg/L and 0.02 mg/L, respectively. The effect of matrices of HALW was negligible under the measurement condition. Repeatability and intermediate precision were 2.5% and 3.1%, respectively. It was successfully applied for measuring Cs in real HALW samples (see table 1). It should be noted that sensitive measurement of Cs by ICP-AES is not possible, while LEP offers a simple technique to measure it.

Table 1: Measurement result of Cs in HALW

Sample	Dilution factor	LEP-OES (g/L)	γ-spectrometry + ORIGEN (g/L)	Correlation
HAW-01	10.000	3.61 ± 0.47	3.60 ± 0.02	good
HAW-02	10.000	0.28 ± 0.04	0.257 ± 0.003	good

Emission lines of Tc in LEP have been found at 254.3 nm (1.0), 261.0 nm (0.87), 264.7 nm (0.5), 363.6 nm (0.18), 371.9 nm (0.11), 403.3 nm (0.15), 423.8 nm (0.06), 426.2 nm (0.1), 429.7 nm (0.15). The figures in parentheses present relative emission intensities considering the emission intensity at 254.3 nm as 1.0. This result indicates that LEP method can be applied to determination of Tc, which is presently in process.

4. Conclusion The analytical method using LEP is very promising in the field of radiochemical analysis. In the study, we have demonstrated its successful applications to measurement of Cs and Tc in HALW samples.

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