Analysis of radioactive iodine-129 by ICP-QQQ using MS/MS mode and an octopole reaction cell with axial acceleration

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APWC2017 in Matsue
POSTER G-10

Introduction

Since the explosion at the Fukushima Daiichi Nuclear Power Plant (FDNPP) resulting from the tsunami in 2011, determination of radionuclides in various samples has been of great interest. Especially important is the analysis of long-lived radionuclides such as iodine-129 (^{129}I: $T_{1/2}$ = 1.57 x 10^7 y) which are measured to investigate the diffusion of radionuclides from the FDNPP, and to study the mechanism of the explosion that occurred at the nuclear reactor.

Isotopic analysis of ^{129}I/^{127}I is challenging because of the enormous signal abundance difference (more than 10^7) between ^{127}I and ^{129}I, both of which must be measured to give a ratio.

In previous work[1)], an Agilent 8800 ICP-QQQ with oxygen (O_2) as the reaction cell gas was used to investigate the possible interference on ^{129}I from ^{127}ID and $^{127}IH_2$. The results indicated that both ^{127}ID and ^{127}IH2 polyatomic ions occur at a level that impacts significantly on the measurement of I at *m/z* = 129. Consequently, blank subtraction of total ^{127}ID and ^{127}IH2 needs to be applied to enable low-level determination of ^{129}I.

When a relatively heavy reaction cell gas such as oxygen is used, the ions passing through the cell lose a lot of energy due to multiple collision with cell gas molecules. As well as causing a significant reduction in analyte sensitivity, the energy loss causes ions to decelerate in the cell,

In the current work, we used the 2nd generation ICP-QQQ, the Agilent 8900, incorporating a new octopole reaction cell with axial acceleration. The 8900 provides a solution to improve sensitivity by allowing an axial acceleration voltage to be applied to the octopole rods. In addition to increasing the transmission and therefore sensitivity of low-energy product ions, axial acceleration also improves discrimination of slow-moving ions that can cause interferences due to artifact signals at the target analyte mass, especially with a relatively heavy cell gas such as O_2. By applying an electric potential gradient in the axial direction, the ions that have been decelerated by collision with the O_2 cell gas molecules can be accelerated again, correcting the loss of sensitivity. The newly developed Octopole cell is shown in Figure 1.

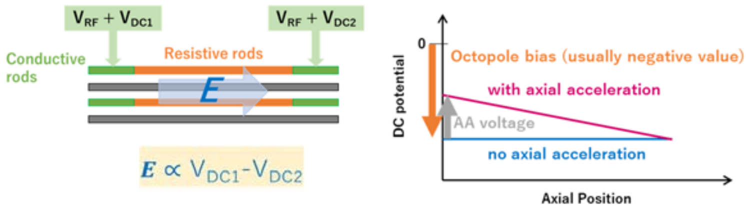

Figure 1 Octopole schematic and potential diagram of Axial Acceleration

In this study, we used ICP-MS/MS with axial acceleration to study the effect of IH_2 and ID interferences on radioactive iodide ^{129}I in more detail. Using ICP-QQQ with O_2 reaction cell gas mode and axial acceleration or deceleration, our results show a significant improvement in the measurable ratio of ^{129}I/^{127}I by in-cell energy discrimination with retarding axial field.

Experimental

❑ **Instrumentation** : ICP-QQQ with MS/MS (Agilent 8900 Triple Quadrupole ICP-MS #100)

Instrumental conditions:

Preset Plasma : Low matrix conditions (RF 1550W, Sampling depth 8mm, Carrier gas flow rate 1.05L/min)

Ion Lens tune : Extract 1 = 0V, Extract 2 = -190V.

ORS and acquisition parameters : O_2 1.05mL/min, He 2mL/min

Axial acceleration or deceleration : -2V to 2V

Results and Discussion

❑ Effect of applied axial acceleration (+ve AA voltage) and deceleration (-ve AA voltage) on the signal at *m/z* 127 (I-127) and 129 (O_2 reaction mode)

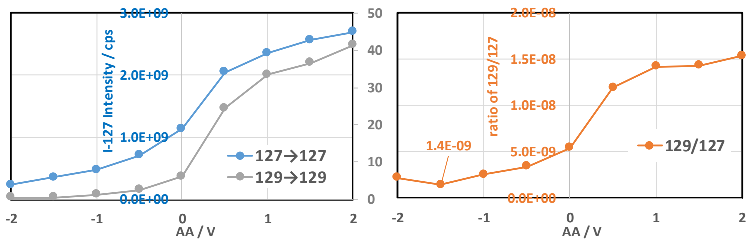

Figure 2 Signal change of ^{127}I & ^{129}I(bkg) (left), and ^{129}I(bkg)/^{127}I (right) vs acceleration voltage.

Compared to the condition of 0 V AA voltage, the signal intensity of ^{127}I was increased by X2.5 with an accelerating AA voltage of +2 V. The background signal at *m/z* 129 also increased with increasing positive axial acceleration voltage. The contribution from I-127 to the signal at *m/z* 129 (ratio of I-129/I-127) increased from 5×10^{-9} to 1.5×10^{-8} with axial acceleration. This may be due to the effective "competition" between AA voltage and KED, which would normally reduce the transmission of polyatomic species of I-127.

❑ Reaction of I^+ and MS/MS setting (O_2 reaction mode)

There may also be a contribution from an endothermic reaction leading to an increase in the background signal from product ions formed in the cell. Production process of IH^+, IH_2^+, ID^+ and MS/MS settings are shown below.

$I^+ + H$	$\rightarrow IH^+$	(Q1: 127 → Q2: 128)
$I^+ + D$	$\rightarrow ID^+$	(Q1: 127 → Q2: 129)
$I^+ + H_2$	$\rightarrow IH_2^+$	(Q1: 127 → Q2: 129)

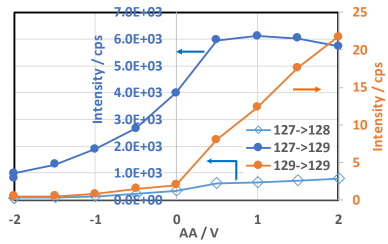

Figure 3 IH^+, IH_2^+ signal change vs axial acceleration voltage

In-cell production of IH^+ and IH_2^+ was confirmed by introducing 100 ppm I-127 solution, as shown in Figure 3 . Increasing the AA voltage appears to promote the generation of these hydride ions. IH_2^+ signal produced was around ten times higher than that of IH^+, although neither of these polyatomics would give a significant signal under normal MS/MS conditions with Q1 at *m/z* 129.

❑ Signal change of ^{127}I, ^{129}I (bkg) for the applied axial acceleration (AA) voltage in the octopole reaction cell by adding He gas as cell gas (O_2+He mode)

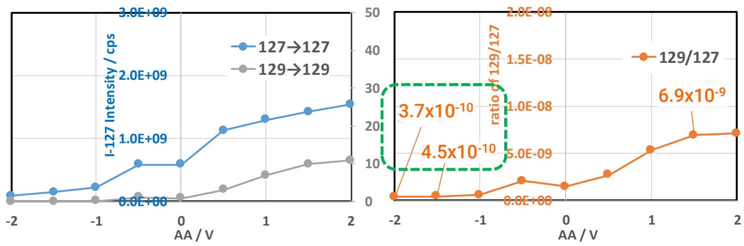

Figure 4 Signal ^{127}I, ^{129}I(bkg) (left) and ^{129}I(bkg)/^{127}I(right) vs axial acceleration voltage.

As with the data shown in Figure 2, both ^{127}I and ^{129}I(bkg) increased with a more positive axial acceleration voltage. As a result, ^{129}I(bkg)/^{127}I was increased and the superior transmission of I-127 polyatomic ions through the cell was assumed. He gas was added (He and O_2 cell gas) in order to attenuate such ions by energy discrimination, and to suppress the formation of cell-formed product ions. The *m/z* 129 background was further reduced by applying a negative axial acceleration voltage, enabling a ^{129}I(bkg)/^{127}I ratio at the 10^{-10} level to be achieved.

❑ Calibration curve for I-129 by O2 on-mass mode with axial acceleration(AA)

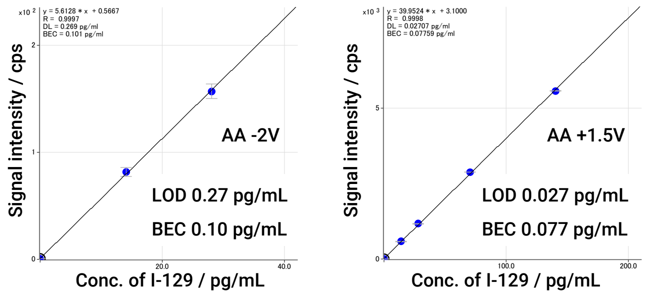

Figure 5 Calibration curve for I-129, O2 cell gas flow 1.50sccm + He 2mL/min with axial acceleration voltage -2V(left) and 1.5V(right).

Conclusions

- By applying a positive axial acceleration voltage in an octopole reaction cell, the sensitivity of $^{127}I^+$ was increased by approximately X2.5, compared with no acceleration.
- However, the generation and/or transmission of I-based polyatomic ions (IH^+, IH_2^+) was also promoted.
- Axial deceleration (negative AA voltage) and the addition of He cell gas was successful in reducing the background signal at *m/z* 129.
- The ratio of I-129/I-127 was decreased to the 10^{-10} level for a sample of 100 ppm I. This background level is an improvement of approximately two orders of magnitude compared with the ratio of 1.3 x 10^{-8} obtained on Agilent 8800 without axial acceleration (reference 2).

References

1) T. Ohno, Y. Muramatsu, Y. Shikamori, C. Toyama, N. Okabe, H. Matsuzaki, J. Anal. At. Spectrom., 2013, 28, 1283-1287.

2) Y. Shikamori, K. Nakano, N. Sugiyama, M. Honda, A. Sakaguchi, K. Sueki, European Winter Conference on Plasma Spectrochemistry, Munster, Germany, Feb 23, 2015 FU2-PO06.