

Improved wide bandwidth mass analysis with MRTOF-MS

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In recent studies of fusion-evaporation reactions using GARIS-II,¹⁾ higher order evaporation channel ($p\alpha n$, $\alpha\alpha n$, *etc.*) products were observed with the multi-reflection time-of-flight mass spectrograph²⁾ (MRTOF-MS). As exemplified in Fig. 1, in many cases, such products had A/q sufficiently different from the neutron evaporation products of primary interest as to make a different number of laps within the MRTOF. As a result, they have a different flight path than the reference ions, which complicates the mass analysis.

We have previously³⁾ developed an analytic framework for making use of such wide mass bandwidth spectra. However, the technique was only accurate on the few-ppm level—sufficient for identification, but insufficient for the precision mass analysis we desire. While the earlier technique was useful at identification of ions within a large mass bandwidth, it was also complicated and required at least two measurements be performed in series under substantially similar conditions, making it prone to errors from *e.g.* slight voltage drifts. Utilizing our presently unique ion preparation trap geometry, we have been able to modify the operation of the MRTOF to allow two measurements to be made nearly in parallel, a scheme we call “concomitant referencing.” This in turn has allowed for development of an improved method to determine the masses of ions within a large mass bandwidth.

In the present implementation of concomitant referencing, the measurement cycle is split into two sub-cycles. Reference ions making different number of laps in each of the sub-cycles are utilized to precisely determine the circulation period, T_{ref} , of the reference ions during the measurement. We then use the original wide mass bandwidth analysis method to determine the number of laps analyte ions made in the MRTOF, and to determine their m/q with sufficient accuracy for identification. Once the number of laps are known, the measured T_{ref} is used to adjust the reference ion ToF, t_{ref} , to match the flight path of the analyte ion:

$$t'_{\text{ref}} = t_{\text{ref}} - \Delta n \cdot T_{\text{ref}}, \quad (1)$$

where Δn is the difference in laps between reference and analyte. Finally, t'_{ref} is used in the single-reference analysis methodology⁵⁾ to determine the analyte ion's

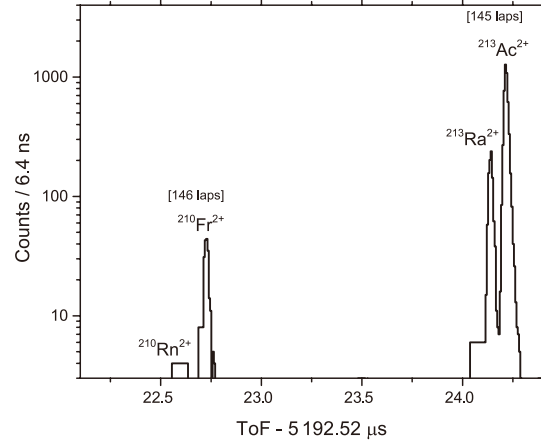


Fig. 1. Portion of online time-of-flight spectrum observed in the study of $^{169}\text{Tm}(^{48}\text{Ca}, X)$ reaction. The $A=213$ isotopes performed 145 laps in the MRTOF, as did the reference ions. The $A=210$ isotopes performed 146 laps in the MRTOF, complicating the mass analysis.

mass-to-charge ratio:

$$m_{\text{analyte}} = m_{\text{ref}} \left(\frac{t_{\text{analyte}} - t_0}{t'_{\text{ref}} - t_0} \right)^2, \quad (2)$$

where t_0 is an inherent delay between the ions leaving the flat ion trap and the start of the TDC, previously⁴⁾ determined to be $t_0=45(5)$ ns.

Offline tests with $^{85,87}\text{Rb}$ have demonstrated an accurate relative mass accuracy of $\frac{\delta m}{m} \sim 10^{-7}$ up to $\Delta n=4$. This makes MRTOF mass spectrometry highly-competitive with storage rings⁶⁻⁸⁾ in terms of mass accuracy and bandwidth.

References

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