



## Full Length Article

# Improving wide-band mass measurements in a multi-reflection time-of-flight mass spectrograph by usage of a concomitant measurement scheme



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## ABSTRACT

We introduce a new concomitant referencing mode for operating a multi-reflection time-of-flight mass spectrograph (MRTOF-MS), wherein the reference and analyte ions are interleaved on a cycle by cycle basis. Using this mode, we demonstrate an improved technique for performing wide bandwidth mass measurements via MRTOF-MS. This new technique offers a simplified analysis and high accuracy.

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## 1. Introduction

The multi-reflection time-of-flight mass spectrograph (MRTOF-MS), first proposed more than 20 years ago [2], is rapidly gaining favor at online radioactive ion (RI) beam facilities, both as an isobar separator and for performing mass measurements [3–7]. These devices are capable of mass resolving powers exceeding  $10^5$  and measurement times on the order of milliseconds [6], while having an improved immunity, as compared to Penning traps, to systematic errors induced by contaminants [8].

While isotope separation on-line (ISOL) facilities can typically deliver low-energy beams of ions within a single isobar chain to an MRTOF-MS, when such devices are connected to a gas cell [9], for use with fusion-evaporation products [10–12] or in-flight fragmentation beams, multiple isobar chains are often extracted simultaneously from the gas cell. Such cocktail beams provide an opportunity for the MRTOF-MS to efficiently utilize online resources by analyzing a great many RI simultaneously. However, as we have previously described [1,8] and will reiterate below, the

multi-reflection nature of the MRTOF-MS results in a non-intuitive  $m/q$  reordering beyond a certain mass bandwidth.

The ability to accurately mass analyze the contents of such cocktail beams as produced by in-flight fragmentation, especially in a simple manner that allows ardent reviewers to recalculate masses from reported data, is highly desirable. As one such example, isotopes believed to participate in the astrophysical r-process are produced at fairly low rates at even the most powerful facilities. However, when produced by in-flight fission and fragmentation, numerous such isotopes can be delivered simultaneously. A technique that could simultaneously analyze the entirety (or even majority) of such a cocktail could reduce the time required to study a broad mass region by an order of magnitude or more.

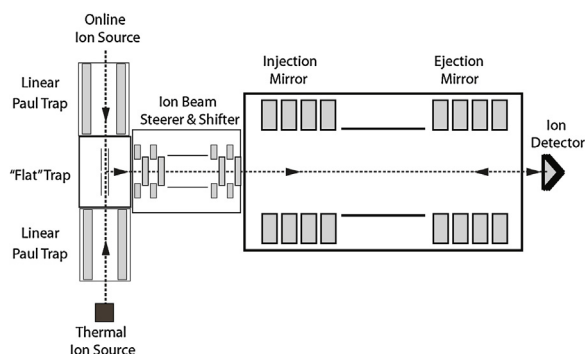
We have previously reported [1] a method to analyze such large mass bandwidth cocktails. Since then, we have made substantive modifications to the apparatus which greatly improve the reliability of mass measurements wherein the reference and analyte make different numbers of reflections in the MRTOF-MS. These include (see Fig. 1) an improved ion trap configuration, the addition of a focusing element and pair of dual ion steerers to improve alignment of the ion beam with the optical axis of the MRTOF-MS, and the implementation of a modified timing system to allow rapid interleaving of reference and analyte ions.

While our earlier technique was useful at identification of ions in a large mass bandwidth, it was also complicated and required at

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**Fig. 1.** Sketch of the MRTOF-MS system. Reference and analyte ions accumulate in the linear Paul traps, and are transferred to the flat trap in an alternating fashion. After cooling in the flat trap, ions are orthogonally ejected toward the MRTOF-MS. A pair of dual ion beam steerers with a focusing element in-between allows the beam to be well-aligned with the optical axis of the MRTOF-MS.

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-MS 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.

This new method bootstraps on the previously developed method. The original method is utilized to determine the  $m/q$  and the number of laps analyte ions make in the MRTOF-MS, with an accuracy of a few parts per million in  $m/q$ . Once the  $m/q$  and number of laps are known, our new method can determine the  $m/q$  more precisely – with a relative mass accuracy of  $\sim 10^{-7}$ . This new operational mode and improved analytical framework makes MRTOF-MS mass spectrometry highly-competitive with storage rings [13–15] in terms of mass accuracy and bandwidth.

## 2. Apparatus

Our implementation of the MRTOF-MS (see Fig. 1) uses a pair of electrostatic ion mirrors, with a single refocusing lens and a long field-free drift region between the mirrors. Fast high-voltage switches are used to lower the potentials applied to the outermost electrodes of each electrostatic mirror in order to allow ions to enter and exit the MRTOF-MS. The device is described in great detail in Ref. [8].

To achieve optimal performance from the MRTOF-MS, ions must be injected as a brilliant ion pulse, with low energy spread, and well-aligned to the MRTOF-MS optical axis. To achieve these requirements, a suite of radio-frequency (RF) ion traps has been implemented, as shown in Figs. 1 and 2a. These RF ion traps are pressurized with  $\sim 10^{-2}$  mbar helium in order to cool ions. At the heart of this ion trap suite is the “flat” trap [16] – a linear Paul trap constructed in a flat geometry. Linear Paul traps are installed on both sides of the flat trap, to accumulate and store ions prior to their transfer to the flat trap. A thermal ion source from HeatWave Labs, installed behind one of the linear Paul traps, provides offline reference ions, primarily  $\text{Na}^+$ ,  $\text{K}^+$ ,  $\text{Rb}^+$ , and  $\text{Cs}^+$ .

The linear Paul traps and the flat trap are each constructed using FR4 printed circuit boards (PCBs) as shown in Fig. 2a. The linear Paul traps were each built from four PCBs arranged in a box configuration. The circuit boards are 135 mm by 8 mm with 15 segments connected by thin film SMD resistors and capacitors on the back side. By capacitively coupling RF signals, with a  $180^\circ$  phase shift between adjacent PCBs, an RF pseudopotential is produced for radial ion confinement. To allow axial manipulation of ions, DC

voltages can be applied at the two outermost electrodes as well as the fourth electrode from the flat trap side. These three DC voltages can be rapidly and simultaneously switched to produce either a DC potential well to axially confine ions or a monotonous axial gradient to transfer ions to the flat trap (see Fig. 2b–e). Reference ions accumulate continuously in one linear Paul trap, while analyte ions continuously accumulate in the other linear Paul trap.

The flat trap is composed of a pair of PCBs mounted on an aluminum support frame. Each of the PCBs has three electrode strips, with the center strip being divided into 7 segments (see Fig. 2a). The flat trap operates in so-called unbalanced mode, where a single-phase RF signal is applied to the outer strips of each PCB to produce a confining radial pseudopotential, while only DC voltages are applied to the segments of the central strip in order to produce an axial potential well. There is a 0.8 mm diameter hole in the middle of the centermost electrode of each PCB; using a pair of switches to rapidly change the voltage applied to the centermost electrode of each PCB, a dipole electric field orthogonal to the plane of the flat trap can be produced to eject ions orthogonally. This capability provides a simple means to accept two separate sources of ions, as well as presenting the opportunity to eject ions in either of two directions.

Between the flat trap and the MRTOF-MS is a pair of dual ion steerers (“Ion Beam Steerer and Shifter” in Fig. 1) with an ersatz Einzel lens in-between them. The recently added second dual steerer and focusing element have improved the alignment of the ion pulse ejected from the flat trap with the optical axis of the MRTOF-MS, surmised from the elimination of lap-number-dependent intensity fluctuations (see Fig. 10 of [8]).

The proper operation of the MRTOF-MS relies on an in-house designed, field-programmable gate array (FPGA) based timing system. This timing system provides timing signals for changing the voltage configurations of the traps (accumulation, cooling, ejection), the configuration of the MRTOF-MS mirrors (injection, storage, ejection), and ejection from the flat trap, as well as setting the RF phase of the flat trap at the moment of ejection.

The times-of-flight of ions passing through the MRTOF-MS are measured with the use of an MCS6A multi-stop time-to-digital converter (TDC) from FAST ComTec. The flat trap ejection triggering signal also serves as the TDC start signal. A MagneToF ion detector from ETP, installed downstream from the MRTOF-MS, provides TDC stop signals when ions strike it after leaving the MRTOF-MS.

The typical measurement cycle for the MRTOF-MS proceeds as follows. First, ions accumulate and cool in the flat trap. Before ions are ejected from the flat trap, the voltage applied to the outermost electrode of the injection-side mirror is reduced to allow ions to pass. In order to give the high-voltage switch sufficient time to settle, this action is performed several microseconds before the ejection of ions from the flat trap. Based on predetermined time-of-flight parameters, the trapping voltage is restored to the injection-side mirror when the ions under analysis are near the turning point of the ejection-side mirror. The ions then reflect between the mirrors for a duration chosen such that ions of a specific  $A/q$  undergo a predetermined number of reflections. After the ions have undergone the desired number of reflections, and while they are near the turning point of the injection-side mirror, the voltage applied to the outermost electrode of the ejection-side mirror is reduced so as to allow the ions to pass. The ions will then travel to the ion detector and produce the TDC stop signals.

## 3. Concomitant referencing

In order to determine analyte ions’ masses from their times-of-flight requires time-of-flight data from reference ions of well-known mass. Ideally, isobaric references would be available

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