High precision mass measurements for the astrophysical r-process

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Abstract. The production of isotopes via the rapid neutron capture process depends sensitively on the atomic mass of the nuclides involved. In light of the recent observation and confirmation of neutron-star mergers as being one of the sites of the r-process, new nuclear data for improved abundance calculations are more urgently needed. To this end, the development of new radioactive ion beam facilities and measurement techniques now extends our reach into unexplored territories. One such facility is the N = 126 beam factory at Argonne National Laboratory. An essential component of this facility will be a multi-reflection time-of-flight mass spectrometer, currently being commissioned at the University of Notre Dame. Mass resolving power of 60,000 using \textsuperscript{39}K\textsuperscript{+} ions has been achieved. Simulation and continued optimization are underway to improve the efficiency of the device.

1. Introduction

Understanding the production of the various elements observed in the Universe has been for a long time a prolific area of research. While the production of the lightest elements via Big Bang, stellar nucleosynthesis, and cosmic ray spallation is mostly understood, the production mechanism of about half of all elements heavier than iron is still under debate. Indeed, beyond the iron peak the various elements are synthesized by three different processes [1]. The slow neutron capture process [2] inside massive stars is responsible for the production of about half of all elements, while the other half comes from the rapid-neutron capture process (rapid since its time scale is on the order of a second), a lesser known process involving very neutron-rich nuclei [3]. The third process, the p-process, accounts for the remaining isotopes.

The r-process is the least known of all nucleosynthesis processes both in terms of its lack of relevant experimental nuclear data and knowledge of its astronomical site. The latter deficiency was mitigated somewhat last year with the first observation of a neutron star merger. It was first observed remarkably in the form of gravitational waves, but its subsequently detected electromagnetic signals were what had significant ramifications for the underlying nuclear physics [4]. The signature transition lines of the lanthanide showed that these heavy elements could be freshly synthesized by the r-process [5]. This important observation essentially confirmed neutron star mergers as being one of the sites of the r-process [6].

The r-process starts from a series of neutron capture from a seed nuclei and proceed until photo-dissociation reaction rates becomes dominant, resulting in increasingly neutron-rich exotic...
isotopes. An equilibrium between the $(n, \gamma)$ and $(\gamma, n)$ reactions along the isotopic chain is then achieved with the only exit point being through the $\beta$-decay of the most exotic isotope in the chain. After this point the series of neutron-captures followed by $\beta$-decays continues in higher-$Z$ elements, forming a path far from stability in the chart of the nuclides that is refereed to as the r-process path.

2. Importance of mass measurements

Under $(n, \gamma)$ and $(\gamma, n)$ equilibrium conditions, the relative abundances of adjacent isotopes, given by the Saha equation, depends exponentially on the one-neutron separation energy $S_n(Z,A+1) = m(Z,A) + m_n - m(Z,A+1)$ where $m$ are atomic masses, and $m_n$ is the mass of one neutron. It is therefore expected that, once established, this equilibrium will only be broken when the neutron supply has been exhausted and the nuclei $\beta$-decay to stability, and so the resulting abundances will be particularly sensitive to atomic mass values.

Unfortunately, the r-process runs far from stability, generally beyond the reach of modern radioactive ion beam facilities (RIB) [7]. Hence there is currently very limited data on atomic masses and one has to rely on nuclear mass models to predict mass values along the r-process path. Commonly used mass models in nuclear astrophysics calculations for the r-process include the Finite-Range Droplet Models (FRDM) [8], Hartree-Fock-Bogoliubov (HFB) approach [9], and Duflo-Zuker [10]. While offering mass values consistent with the 2016 Atomic Mass Evaluation (AME2016) [11] and with one-another close to stability, these models diverge far from stability, in the region of the r-process path [12, 13]. These conflicting mass predictions results in variations in the calculated abundance pattern [14, 13].

Over the past few years, increasingly elaborate sensitivity studies on the effect of atomic masses on the abundance pattern produced by the r-process have been performed [15, 16, 17]. This was studied using different mass models, as well as different astrophysical conditions [13]. The sensitivity of other experimental quantities, including neutron capture rates, beta decay rates, and beta-delayed neutron emission probabilities were also undertaken [13]. Among all these various quantities atomic masses were found to have the greatest impact [13]. The latest sensitivity studies were fully self-consistent, where the variation of the uncertainty on one atomic mass was propagated to the other quantities since atomic masses enters in the determination of all of them [17]. As a result of these studies several conclusions can be drawn [13]: first, variations of the atomic mass of a single nuclei by a value as low as 100 keV can have an impact on the abundance pattern in general. Second, the most influential nuclei are located near the closed $N = 82$ and 126 shells as well as the rare-earth region located around $A = 165$. Finally, while the abundance pattern is sensitive on the choice of mass model or astrophysical condition, similar nuclei appear as being the most sensitive.

To minimize the uncertainty in the abundance pattern associated with a single atomic mass, ideally the measurement should be performed down to an uncertainty of less than 100 keV, which for the mass region where the r-process takes place corresponds to a relative uncertainty of less than $\delta m/m \sim 10^{-7}$.

3. The accuracy of Penning trap mass spectrometers

The field of precision atomic mass spectrometry can be divided broadly by their uses of indirect and direct techniques [18]. Indirect techniques involve the measurement of reaction or decay $Q$-values, which related to a difference between several masses. On the other hand, in direct techniques only two nuclei are measured: the nuclei of interest and a calibrant (or reference). These direct techniques can be further divided in two groups: time-of-flight spectrometry and frequency measurements [18]. Each mass spectrometry techniques offers its
own advantages. For example, single-turn time-of-flight measurements can accommodate the shortest-lived nuclei and nuclei produced at very low yield. But the direct measurements of cyclotron frequency like those done in Penning traps allow for the most precise mass determination, where relative uncertainties on the order of $\delta m/m \sim 10^{-7}$ are routinely achieved [12]. Penning traps are, however, limited to nuclei with half-lives of 100 ms or more and are not accommodating to high levels of contamination of the incoming beam. It should be noted that nuclei with half-lives as short as 8.8 ms have been measured with a Penning trap [19].

It is not sufficient for the atomic mass measurement to be only precise; they also needs to be accurate. Ultimately, one can have confidence in the accuracy of the mass of a nuclei only if it has been measured more than one time, and by different experiments, employing various techniques like those described above, which will have different technique-dependent systematics. Despite being by far the most precise measurement technique available, therefore making its measurements difficult to compare with others, we must still test the consistency of the atomic mass of a given nuclei that has been measured through repeated measurements.

![Figure 1.](image)

Figure 1 illustrate the consistency of measurement performed using Penning traps. This figure shows the relative deviation of a given mass measurement of a certain isotope versus the most precise Penning trap measurement of that isotope. Essentially, we define the relative deviation $\Delta R_i$ as:

$$\Delta R_i = \frac{m_i(N,Z) - m_0(N,Z)}{\delta m_i(N,Z)},$$

where $m_i(N,Z)$ is a given mass measurement of a certain isotope, $m_0(N,Z)$ is the most precise measurement of that isotope, and $\delta m_i(N,Z)$ is the uncertainty on the measurement of the isotope of mass $m_i(N,Z)$. Hence $\Delta R_i$ is a measure of how many standard deviations a certain measurement is from the most precise measurement. The values of $m_i(N,Z)$ and $m_0(N,Z)$ where calculated directly from the input values of AME2016 [11]. As shown in Figure 1, most mass measurement are within one standard deviation. If all measurements, including the ones that were measured multiple times by the same group (often times as part of systematic studies), are considered and fitted with a Gaussian distribution, a standard deviation of 0.74 is obtained. The standard deviation is lower than one primarily due to the large number of measurements that were measured by one group multiple times, giving relative deviations close to zero. This experimental setup-related bias in the determination of the consistency of Penning trap measurements can be removed by considering only the isotopes that were measured by more than one experimental setup. The values of $\Delta R_i$ shown in dark color in Figure 1 only consider the masses $m_i(N,Z)$ and $m_0(N,Z)$ measured by different groups. The resulting distribution is more Gaussian and with a standard deviation of 0.88; closer to one. This comparison shows that, in general, measurements performed multiple times by the same experimental setup and different experiment yield consistent values with a nearly statistical spread.
4. Penning trap mass spectrometers for the r-process

There are currently seven operational Penning trap mass spectrometer located at RIB facilities in Europe (ISOLTRAP at CERN [20], SHIPTRAP at GSI [21], JYFLTRAP at the University of Jyvaskyla [22], and TRIGATRAP at the TRIGA reactor [23]) and North America (CPT at ANL [24], LEBIT at the NSCL [25], and TITAN at TRIUMF [26]). They are located at facilities with complementary reach in terms of the types of isotope produced and the properties of the beam. Fission so far has been the reaction of choice to produce nuclei of relevance for the r-process since the fission distribution is located around the $N = 82$ shell and the rare-earth region. The JYFLTRAP and CPT systems have been particularly active, with their measurements covering a large portion of the $N = 82$ shell and the rare-earth region [27, 28, 29, 30]. This is because the production method of the radioactive ion beams at their facilities (IGISOL [31] for JYFLTRAP and CARIBU [32] for CPT) is element-independent. While the production mechanism of the radioactive nuclei is different at these two facilities (proton-induced fission on uranium at IGISOL and the spontaneous fission of $^{252}$Cf at CARIBU), both of them the radioactive nuclei recoil out of the material (target or source) in an ionized form and allowed to thermalize in a helium gas volume. This is in contrast to ISOL facilities [33], where the radioactive nuclei produced in the fission diffuse out of the target only when a high heat is applied. The diffusion process is element specific and typically it is difficult to get refractory element out in sufficient quantities. It should, however, be noted that a small amount of the radioactive refractory $^{54}$V was observed at TITAN [34]. Nevertheless, ISOL facilities have their advantages and sometimes surpass thin-target facilities; their thick targets provide greater opportunity for reactions to occur, and therefore more nuclei of interest can be produced provided they can quickly diffuse out and be ionized. For example, the TITAN Penning trap at ISAC, and ISOLTRAP at ISOLDE, measured very neutron-rich Sr-Rb isotopes [35, 36, 37, 38] of interest for the r-process, as well as neutron-rich Cd [39, 40] and In [41] isotopes more recently.

5. Production of critical r-process nuclei at future RIB facilities

Looking in the future, new large user facilities such as FRIB [42] and FAIR [43] will provide more neutron-rich RIBs approaching, and in some cases reaching the r-process path. As an example, Figure 2 shows the mass-measurement reach of the future FRIB facility together with the most sensitive masses for a high-entropy hot wind r-process environment [13]. On this figure, the greater the value of the sensitivity parameter $F_{\text{max}}$, the more sensitive is the r-process abundance pattern to that particular atomic mass. Hence, as the figure indicates, a large fraction of critical masses for the r-process would be accessible to the mass spectrometers located at FRIB.

Before the advent of next-generation large-user facilities, one isotopic region remained stubbornly out of reach: $N = 126$ shell closure. This region is of interest for the r-process since it is related to the production of the last abundance peak and, as can be seen in Figure 2, mass uncertainties here have high impact on final abundances. Unfortunately it falls far beyond the two fission distributions of $^{235}$U, and even projectile fragmentation has trouble producing sufficiently exotic nuclei in this region [44]. Fortunately, one can access more neutron-rich in this region by using deep-inelastic reactions [45], where nucleons are exchanged in the near-miss of two stable, similarly heavy neutron-rich species. The KEK Isotope Separation System (KISS) facility at RIKEN [46] is currently employing such a reaction to produce beams near the $N = 126$ shell closure. A second facility using that type of reactions is currently under construction at ANL. Called the $N = 126$ beam factory, it will impinge a $^{136}$Xe beams at 9-10 MeV/u from ATLAS on a $^{198}$Pt target. But here, again, contamination by nearby species remains high, and several types of experiments, such as mass and half-lives measurements, require contaminant-
The capture and release of the bunches can be accomplished either by switching the ion source off and on or by applying a pulse of high voltage to the electrodes. The bunches of interest can then be accelerated, collimated, and focused by apertures to achieve high brightness, allowing them to be injected into the MR-ToF. In the MR-ToF, the ion bunches pass through two electrostatic mirrors, each comprising five electrodes. The mirrors are designed to have a different assembly design compared to the ISOLTRAP MR-ToF [48]. A schematic of the MR-ToF is shown in Figure 3. It consists of two electrostatic mirrors, each comprising five electrodes. These mirrors are separated by a gap of a small enough width to allow the passage of the ions but not the passage of electrons. The working principle of the MR-ToF is fairly simple: a cocktail bunch containing several different isotopes is captured in the MR-ToF and allowed to bounce back-and-forth between the two mirrors. The various isotopes will start to be resolved according to their mass-to-charge ratios, forming distinct groups. After a certain time, the bunch is released from the trap and contaminant ions can be separated by their time-of-flight from the ion of interest. The capture and release of the bunches can be accomplished either by switching the potential of the entrance and exit side mirrors, or by switching the potential on the central drift tube relative to the mirrors [49]. Due to its simplicity, and with the advantage of leaving more electrodes static, the second method was chosen for the Notre Dame MR-ToF.

The off-line test setup, shown on Figure 3, comprises an alkali ion source producing all naturally occurring alkali isotopes from $^{23}$Na to $^{133}$Cs. After the ion source, a Bradbury-Nielsen gate (BNG) is used to chop the continuous beam into short bunches down to 60 ns in length. After, Einzel lenses and steerers are used to focus and align the bunches before injection into the MR-ToF. After the MR-ToF, a second Einzel lens is used to focus the beam on a MCP. Before and after the MR-ToF, Faraday cups and MCPs are placed on actuators such that they can be put in place for diagnosis. The whole system is pumped by four 300 L/s turbo pumps backed
by a scroll pump. A vacuum around $2 \times 10^{-10}$ mbar is typically achieved at the location of the MR-ToF.

**Figure 3.** Top: Schematic of the beam line for the off-line commissioning of the ND MR-ToF. Bottom left: Time-of-flight spectra of a bunch created by switching the BNG and reaching the last MCP (without being captured). Bottom right: Photo of the beam line at the University of Notre Dame.

**Figure 4.** Variation of the mass resolving power, $R$, and the full-width half maximum of $^{39}$K$^+$ bunches as function of the number of round trips.

Figure 4 shows that a mass resolving power reaching 60,000 has been achieved after 200 round trips which is sufficient to resolve isobars. The large increase and peaking of the resolving power at 200 round trip is a consequence of having the time-focus placed at that number of round trips. While the targeted minimum mass resolving power has been reached, the efficiency of the device needs to be improved. To that end, a second steerer has been installed between the BNG and the first Einzel lens. Simulation results show that 100% of the ions from the BNG can be made to reach second MCP through lensing now done with this new steerer, a factor of ten increase compared to the previous configuration. More simulations in conjunction with optimization in the laboratory are currently underway.
7. Conclusion

More nuclear physics inputs are needed to better understand the astrophysical r-process (even more critical after the neutron star merger event observation). Atomic masses in particular are one of the most important nuclear data input for r-process abundance calculations. Penning traps are the most precise and accurate mass measurement method to date. Several new masses at Penning traps worldwide have been measured, improving our understanding of the r-process. The last r-process abundance peak around N = 126 is the one with the least experimental information available. The ANL N = 126 beam factory will produce critical nuclei for our understanding of the r-process for the first time and the ND MR-ToF will be used there to remove isobaric contamination from the beam. The MR-ToF is currently being commissioned off-line at Notre Dame and mass resolving powers reaching 60,000 have been observed. Means to improve or understand efficiency losses are being investigated.

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