Production of Radioactive Molecular Ions in Radiofrequency Quadrupole Gas-Reaction Cells

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Abstract. Limited types of radioactive molecules (RM) can be made inside hot-cavity targets at ISOL facilities like TRIUMF. However, extreme conditions in these targets present formidable unsolved challenges to efficient production and delivery of RM's. Here we propose using RFQ gas-reaction cells to produce RM from radioactive ion beams (RIB) by room temperature RIB-gas chemical reactions at eV energies. Two options are possible: (1) using an ion reaction cell (IRC) that is a linear RFQ ion guide and reaction cell used as an 'on-line ion source', and (2) using the ARIEL RFQ cooler-buncher (ARQB). RFQ gas-cells are a controllable and efficient method to produce RM from chemical reactants that cannot be used in ISOL targets. This 'on-line chemistry' offers a way to enable groundbreaking Beyond Standard Model (BSM) physics research, using a wide diversity of new rare and exotic RM beams that would be difficult or impossible to produce in hot-cavity targets.

1. Introduction

Radioactive molecules (RM) including RaF_x, $Ra(OH)_x$, RaH_x, ThO_x, BaH_x, YbO are exotic molecular species containing rare radioisotopes. These RM decay spontaneously, dissipating energy by the emission of radiation. Isotope separation on-line (ISOL) facilities like TRIUMF and CERN-ISOLDE synthesize a limited set of molecular species alongside radioactive ion beams (RIB) in their hot-cavity target ion sources [1, 2, 3, 4]. The RM form in cavity targets and extraction systems by a complex and poorly understood cascade of reaction kinematics following fission, spallation and fragmentation after the primary p^+ beam impinges on solid targets containing elements such as Si, Ta, Nb or U in the form of metal foils or composite ceramics.

Next-generation subatomic experiments wish to use RM in new Beyond Standard Model (BSM) physics searches, i.e. charge (C), parity (P) and time (T) symmetry violation effects in

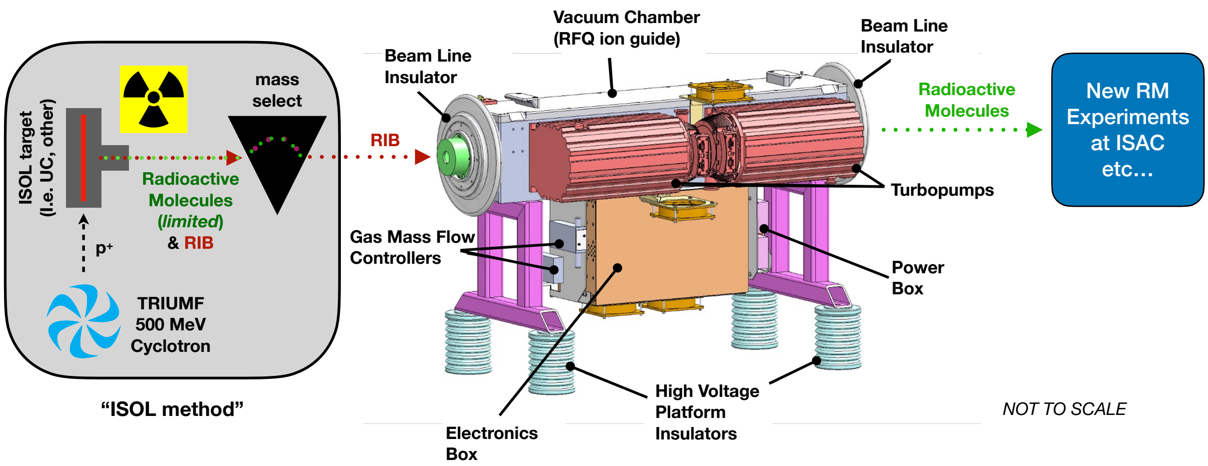

Figure 1. Sketch of the proposed RM production setup at TRIUMF, using an ion reaction cell (IRC). Mass-selected RIB from the ISOL target enter the IRC and undergo controlled ion-gas chemical reactions inside the RFQ reaction cell. Product molecules exit the IRC to downstream experiments. A model of the IRC is shown (*reproduced with permission from Isobarex Corporation*).

molecules, implications for dark matter, and fundamental topics in quantum chemistry [1]. These studies require efficient RM production of the required molecule at high purity and yield. However, the extreme conditions in hot-cavity on-line targets (e.g. high temperatures, poor vacuum, high radiation fields, isobaric interferences, unfavourable chemical conditions and reaction energies) present a major challenge to targeted molecule production for experiments.

RM creation from RIB in radiofrequency quadrupole (RFQ) ion guides by ion-gas chemical reactions at room temperatures is a more versatile, straightforward and efficient technique that allows controlling reaction conditions as well as reactants. Molecules containing radioactive isotopes are known to form unintentionally in cooler-bunchers [12]. However, in those cases, undesired charge exchange with the primary radioactive beam occurs. The rich array of ion-gas reaction chemistries in RFQ's to make and destroy molecules are very well established [6, 7] but remain unexploited for subatomic physics. Here we discuss the idea of using RFQ gas-reaction cells at ISOL facilities as an efficient and robust method for producing RM by 'on-line' RIB-gas chemistry.

2. Production of Molecules in RFQ Gas-Cells

Presently at TRIUMF, RIB are produced at the ISAC (Isotope Separator and ACcelerator) facility by the ISOL method where protons accelerated by the main cyclotron (up to 520 MeV at 100 μA) impinge on solid targets (see Fig 1). By 2026, ARIEL (the Advanced Rare IsotopE Laboratory) will add one additional p^+ target, and one novel 50 MeV electron photofission target to produce rare neutron-rich isotopes [8, 9].

To fully exploit ARIEL and ISAC capabilities, two RFQ systems may be considered for RM production. Fig 1 and Fig 2(a) shows the ion reaction cell (IRC), while Fig 2(b) shows an internal schematic of the ARIEL radiofrequency quadrupole cooler-buncher (ARQB).

2.1. Ion Reaction Cell (IRC)

The IRC in Fig 1 and Fig 2(a) is a room-temperature gas-filled RFQ ion guide and reaction cell that can accept positive or negative atomic ion beams at ISAC energies up to 40 keV (currently, only positive ions are extracted from ISAC targets). Molecules are produced (or destroyed) at low eV energies inside the RFQ gas cell from a wide variety of pure gaseous reactants under controlled conditions [5]. Product molecules are finally delivered from the instrument exit port to experiments. Fig 1 shows the overall IRC construction, including vacuum system components, the chemically resistant mass flow controllers, PLC and HV hardware, and a conceptual integration into TRIUMF radioactive beamlines. Fig 2(a) shows the IRC internals, including the optics at beam entry (deceleration), the RFQ ion guide and reaction cell where molecules are produced by ion-molecule interactions with gases or vapours, and optics for beam exit (reacceleration).

The IRC column in Fig 2(a) contains 13 RFQ segments made from chemically resistant Ultem, modular and reconfigurable. DC voltages are applied to individual rod segments and apertures. This flexibility means the gas reaction cell is alterable and can be tailored, e.g. cell volume and length, gas pressure and number of collisions, use of multiple reaction gases, collisional cooling and energy distribution of ions along the RFQ column. Fine control over the energy transferred to the ions is possible, for example, when forcing endothermic reactions to proceed. Efficient optimization of molecule production may then occur at eV energies by highly selective chemical reactions, where formation of other undesired molecules is minimized. Molecule production rather than destruction is achieved by providing favourable energy conditions for a suitable reactive gas with an electronegativity close to the ion of interest. This promotes chemical bond formation for a large diversity of molecules in high-yields and high-purities from many chemical reactants that could not be used inside hot-cavity on-line targets.

Typical operating parameters in negative ion mode used by [5] to induce ion-gas chemical reactions were 5 mTorr gas pressure at room temperatures and $V_{DC\ rods} = 14$ V, with $V_{ion\ source} = -20931.2$ V, $V_{IRC\ deck} = -20924.2$ for a 7 V difference. Analyte ions had the opportunity to experience at least several collisions. Simulations are now being performed for positive ions at ISAC, to determine the optimal time in the cell for a reaction to occur, required cell length, energy profiles of the ions, and gas mixtures to simultaneously collisionally cool and promote chemical reactions.

The system rests on four 40 kV high voltage insulators allowing the kinetic energy of the incoming atomic ions to be adjusted within <1 eV of the appropriate energy to react with gases introduced to the cell. The RFQ column is removable so that the empty vacuum box can be used telescopically to transfer beam between the entry and exit Einzel lenses without the column in place. The RFQ column has a current limit of $\sim 1\ \mu$A. Electronics on the high voltage platform are connected to computer control at ground by a 5G wi-fi or fibre optic link. Reaction gases are introduced separately as required.

Initial setup and tests of the IRC will occur with stable-isotope beams at TRIUMF's ISAC Test Stand Facility (ISTF). Here, stable molecule production will be quantified with a diverse variety of gaseous inorganic and organic compounds reacting with positive or negative stable atomic ion beams (ISAC uses only positive beams, but negative beams can be implemented). This will (i) establish a clear understanding of stable molecular reactions that are relevant to future RM production from RIB, and (ii) offer research opportunities to both subatomic physics and broader scientific problems. Elements of interest include Al, Ca, Ba, Mn, Fe, Rb, I, W, Mo, Ti, Cr, Cs, Re, Pb, La, U. Lanthanides and actinides are of special interest. We aim to systematically study the kinetics of the formation of molecules for hydrides, oxides, fluorides, carbides, hydroxides, chlorides and other more complex molecules, i.e. polyatomic, heavy element and dia/paramagnetic molecules, and use these data to quantify IRC performance. These molecules are of high interest to RM subatomic experiments and theory. Following successful tests and further system refinement, the IRC will be moved to ISAC for long-term integration with the radioactive beam systems for RM production from RIB.

The IRC system is being developed by a collaborative TRIUMF-industry partnership. Developments are being explored to deliver bunched beams from the system. This method of rare molecule production and preparation offers a significant advancement for fundamental particle

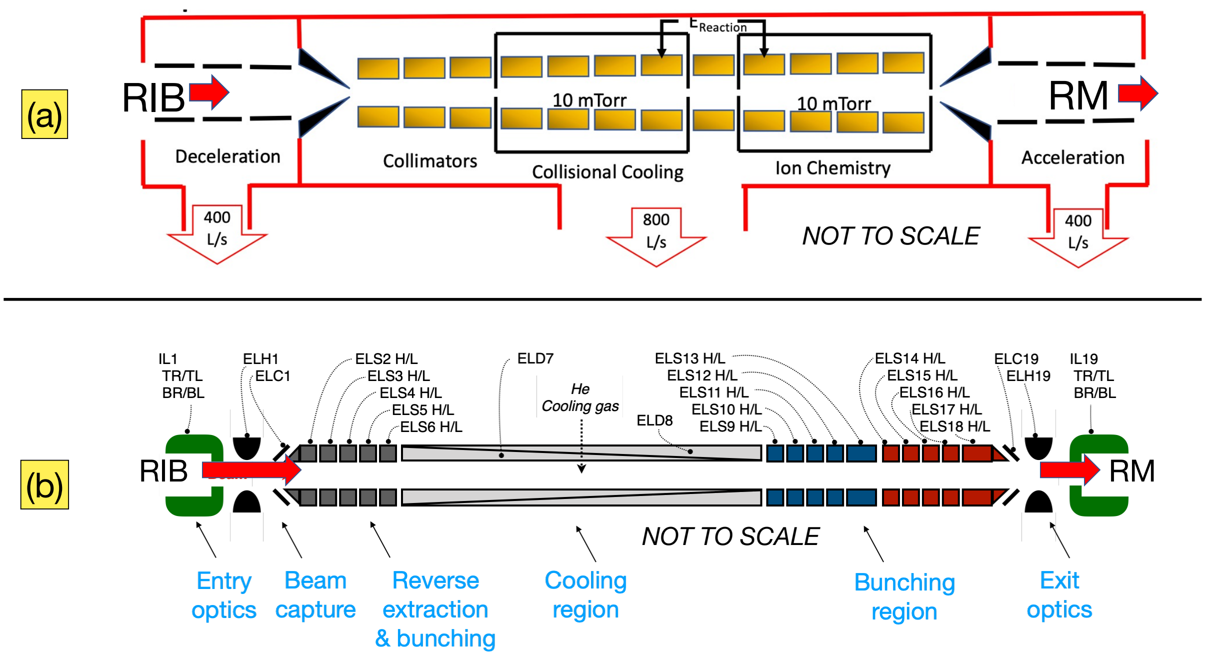

Figure 2. Internal sketches of (a) the ion reaction cell (IRC) and (b) the ARIEL RFQ cooler-buncher (ARQB).

and nuclear physics with molecules, and strengthens the science opportunities for upcoming low-energy precision experiments at TRIUMF.

2.2. ARIEL Radiofrequency Quadrupole Cooler-Buncher (ARQB)

CANREB (the CANadian Rare isotope facility with Electron Beam ion source) is dedicated to preparing and delivering beams of highly charged ions (HCI) for experiments. CANREB is part of the larger ARIEL facility [8, 9] and includes the ARQB schematically shown in Fig 2(b) [10]. The ARQB cools and bunches RIB spanning the mass range up to U at $\leq$100 Hz repetition rates containing $< 10^7$ ions/bunch. Bunch energies are then adjusted by a pulsed drift tube to $6 < E_{bunch} < 14$ keV immediately before acceptance into an electron beam ion source (EBIS) for charge breeding [11].

Although the main purpose of the ARQB is to prepare RIB for charge-breeding, RM production in a similar manner to the IRC should be possible in the ion guide. Three advantages of the ARQB over the IRC are that (1) the system was commissioned as of 2021 summer, now allowing RIB from ISAC to be used to produce RM, (2) beam bunching is possible, and (3) options exist for cryogenic cooling of He buffer gas to further reduce beam emittance. However, two major disadvantages are that (a) the ARQB is not designed for gas-reaction chemistry, particularly with reactive chemicals including F_2, Cl_2, Br_2, H_2O, alcohols and other difficult compounds, and (b) cryogenic operation will limit the range of reactants due to freeze-out.

We are exploring possibilities of using the ARQB for RM production with a range of reaction gases. Ion optical simulations are also being developed to determine how best to extract RM beams from CANREB. Options include 'bouncing' out of the EBIS (without electron beam), deflecting off electrostatic lenses before the EBIS, or using the ARQB in reverse-extraction mode after RM formation.

If RM extraction can occur towards the EBIS, mass analysis may be possible by the Nier spectrometer (NIS) [10] before RM injection into ISAC for experiments. However, CANREB beamlines from the NIS can only go to 2 places: the ARIEL yield station, or the ISAC-I linac that cannot accelerate heavy molecules. Therefore, sending RM to the ISAC low energy area will need additional beamline infrastructure to deliver for a low-energy experimental program. The case for reverse-extraction from the ARQB will similarly require beamline infrastructure to deliver to ISAC-I. Options are being considered how to deliver RM to experimenters once the controlled formation of molecules inside both the ARQB and IRC are understood experimentally.

3. Summary

RFQ gas-reaction cells are a robust, simple method for RM production rather than inside ISOL targets where non-equilibrium, high temperature conditions exist in an environment abundant with reactants from all elements and isotopes produced in the target by spallation, fragmentation and fission (actinide targets). Currently, no other laboratory has a dedicated rare RM beam facility. Such a facility at TRIUMF has the potential to (1) significantly expand the portfolio of available beams in the ARIEL operation era for research, (2) strengthen the science opportunities for low-energy precision experiments on radioactive molecules, (3) offer many interdisciplinary opportunities for students, and (4) develop new scientific collaborations with external partners.

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