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A setup to develop novel Chemical Isobaric SEparation (CISE)

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ABSTRACT

Gas catchers are widely used to thermalize nuclear reaction products and subsequently extract them for precision measurements. However, impurities in the inert stopping gas can chemically react with the ions and thus influence the extraction efficiency. So far, chemical reactions in the gas-catcher have not been investigated in detail. Therefore, we are currently building a new setup to develop Chemical Isobaric SEparation (CISE) with the aim to understand the chemistry inside the gas-catcher and to explore its potential as a new technique for separation of isobars. In this paper, we give a short description of the setup together with the ion transportation studies performed via ion-optics simulations.

1. Introduction

Experimental studies of nuclei far away from the valley of stability are essential for further development in the field of nuclear structure and nuclear astrophysics. A main challenge in these experiments are the low production yields of the nuclei of interest along with the formation of a significant amount of unwanted by-products. This sets constraints on, e.g., high-precision mass measurements of exotic nuclei. A large fraction of non-isobaric by-products can be suppressed in an electromagnetic separator behind the reaction target. However, the separation of isobaric contaminants in most cases is challenging and an additional separation step is required. We intend to employ a novel chemical separation technique behind a recoil separator to select isobaric nuclides through their chemical properties. The chemical reactions will take place within a gas-catcher device used for stopping and thermalizing the high-energetic recoils [1–4]. Generally, the aim is to keep the level of impurities in the inert gas as low as possible in order to avoid the loss of ions through the formation of molecules or charge-exchange reactions. The quest for purity is one of the reasons why the new generation gas catchers are often operated at cryogenic temperatures [2,3]. However, in a small number of experiments, gas-chemical reactions have been used to separate the nuclide of interest from unwanted background [5]. The full potential of chemistry in a gas catcher has not been

explored, yet. Therefore, we are building a setup consisting of a gas catcher at room temperature into which we will introduce chemical reactive gases in addition to helium. In order to separate the ions of interest we aim at establishing a suitable chemical system which, ideally, would allow background isobars to react with the chemical reagent forming charged molecules and/or neutral atoms, while the ions of interest are unaffected. A potential chemical reagent to study, e.g., nuclei in the ¹⁰⁰Sn region is methane [6].

2. The CISE setup

To investigate the capability of isobaric separation, the new CISE (Chemical Isobaric SEparation) setup is under construction. The layout of the setup is shown in Fig. 1. It consists of a gas-catcher and an ion-guide, which is coupled to a commercial quadrupole Time-of-Flight (qToF) spectrometer. The CISE setup will be used in offline experiments, in which chemical reactions with stable ions produced by laser ablation will be investigated, as well as in online experiments with short-lived isotopes produced in nuclear fusion reactions.

2.1. The gas-catcher

The size of the gas-catcher is optimized to thermalize fusion-

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Fig. 1. Schematic of the CISE setup consisting of a gas catcher and an ion guide that are coupled to a commercial qToF. The ion-guide system consists of Hexapole 1 and Hexapole 2. The qToF is a quadrupole mass spectrometer (Quadrupole MS) coupled with a reflection Time-of-Flight mass spectrometer (ToF MS).

evaporation residues in the mass region around $A = 100$. The stopping distribution of ^{100}Sn with a kinetic energy of 67 MeV has been simulated using SRIM [7]. As a stopping medium a 6.5 μm thick titanium entrance window and a mixture of 95% of He and 5% of CH_4 at 50 mbar was considered in the simulation. Taking into account the stopping efficiency and the extraction time, the size of the gas-catcher chamber was fixed to 450 mm in length and 400 mm in diameter. The gas catcher is designed in a way that it can be easily divided by an additional RF carpet into two subsequent sections to avoid problems through plasma formation or molecular break-up. In the first sections, the ions will be slowed down in helium, in the following section reactive gases will be introduced.

The main components of the gas catcher are shown in Fig. 2. There are two target holders implemented in the setup to produce ions of stable isotopes by laser ablation. The ions are guided along the extraction axis by a Direct Current (DC) gradient, which is applied to the ring electrodes. At the backside of the catcher is an RF carpet, a printed-circuit board with concentric ring electrodes and an electrode gap of 0.125 mm. The ions are guided to the center of the carpet through a DC gradient and a Radio-Frequency (RF) potential, which are applied to the electrodes. From the center, the ions are extracted through an exit hole of 0.45 mm diameter by a supersonic gas flow.

The ion trajectories along the electrical fields were simulated using

Fig. 2. The technical design of the gas catcher and the coupling ion guide.

Fig. 3. Ion trajectories in the vicinity of the exit hole of the RF carpet. The color code refers to the kinetic energy of the ions. The simulation includes gas flow as well as DC and RF field calculations. The collisions of ions with helium gas are simulated based on viscous-drag model. Simulated transmission is 100% and the phase space distribution 2 mm from the back side of the RF carpet is shown in Fig. 4(a). (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

the COMSOL Multiphysics™ modeling software and are shown in Fig. 3 as they approach the exit hole of the RF carpet. In the gas-flow calculation, we assume a pressure of 50 mbar inside the gas-catcher and roughly 0.01 mbar in the hexapole section close to the hole. The DC gradient for the DC ring electrodes is set to 7 V/cm and for the RF carpet to 3 V/cm. The RF potential with an amplitude of 100 V_{pp} and a frequency of 10 MHz is applied to the RF carpet electrodes in a way that the RF phase of the neighboring electrodes differs by 180°.

2.2. Coupling of the gas catcher to the qToF

From the gas catcher, the ions are carried by the supersonic gas jet with a cone shape distribution (see Fig. 3)) into the double hexapole ion-guide section. The first hexapole is placed 2 mm behind the RF carpet. It is separated through a diaphragm from the second hexapole. Each hexapole consists of six stainless steel rods of 5 mm diameter. The distance between centers of two opposite rods is 13.9 mm and an RF voltage with 120 V_{pp} at 1 MHz frequency is applied. Through differential pumping, the pressure is reduced from 4×10^{-3} mbar in the area of the first hexapole to 6×10^{-5} mbar in the second hexapole chamber.

Fig. 4. Ion trajectory simulations using the COMSOL Multiphysics software: a) results of the simulation for the phase space of the extracted ions (Ag^+ and AgCH_4^+) from the gas catcher at 2 mm distance behind the RF carpet; b) phase space of the ions at the end of the second hexapole; c) energy spectrum of the ions, yellow (cross pattern): extracted ions from the gas catcher with 0.062 eV FWHM. Blue (solid pattern) and purple (diagonal pattern): energy spectrum of the atomic and molecular ions both with 0.7 eV FWHM energy spread at the end of the first hexapole; d) energy spectrum of the ions when they reach the end of the second hexapole (FWHM = 2.5 eV for atomic ions and 2.35 eV for molecular ions); e) ion trajectory simulations of the ions in the double hexapole assembly. (For interpretation of the references to colour in this figure legend, the reader is referred to the web version of this article.)

The first hexapole has a length of 125 mm, which according to simulations in COMSOL is short enough to prevent major losses of the ions due to collisions with the gas. The second hexapole has a length of 550 mm and is directly coupled to the qToF.

The transport of atomic ions (e.g. Ag^+) as well as molecular ions such as AgCH_4^+ through the ion-guide system have been simulated by using the COMSOL multiphysics software. Fig. 4(a) shows the phase space of the extracted ions from the gas catcher at a distance of 2 mm behind the RF carpet where the ions enter the short hexapole. The phase space of the ions evolves by passing through the ion-guide assembly; the correlation between velocity and position of ions is lost at the end of the ion-guide assembly (b). The yellow histogram (cross pattern) in Fig. 4(c) shows the energy of the extracted ions at the beginning of the ion guide. The ions lose energy by collision with the buffer gas in the first hexapole with a pressure of 4×10^{-3} mbar, which in turn leads to a significant broadening of the energy spectrum. The energy spectrum can even extend to energies higher than the initial energy of the extracted ions due to the RF heating of ions in the hexapole. The blue (solid pattern) and purple (diagonal pattern) histograms show the energy spectrum of atomic and molecular ions at the end of the first hexapole. The energy loss of the molecular ions is high compared to the atomic ions due to the larger geometrical collision cross section with buffer gas atoms. The collision cross sections of atomic ions Ag^+ and of molecular ions AgCH_4^+ with the buffer gas

(mixture of 95% He + 5% CH_4) are $\sigma = 3.12 \times 10^{-19} \text{ m}^2$ and $8.33 \times 10^{-19} \text{ m}^2$ according to the following equation:

$$\sigma = \pi (R_{\text{ion}} + R_{\text{He}})^2 \times [0.95] + \pi (R_{\text{ion}} + R_{\text{CH}_4})^2 \times [0.05] \quad (1)$$

where R_{ion} , R_{He} and R_{CH_4} are the van der Waals radii of the ions and the atoms of the buffer gas mixture.

A potential difference of 14 V between the first and the second hexapole assures the acceleration of the ions towards the qToF. Fig. 4(d) shows the energy spectrum of ions at the end of the hexapole doublet. The ion trajectories in the ion-guide assembly are shown in Fig. 4(e). According to the ion optical simulations, the overall transmission efficiency of the ions through the hexapole ion-guide assembly is above 90% for both atomic and molecular ions.

From the ion guide, the ions are injected into a commercial qToF 2 from Micromass UK Limited. The qToF consists of a quadrupole which can be used either as a mass spectrometer for ions of mass from 10 to 4000 Dalton (mass-to-charge ratio) with a mass resolving power $m/\delta m = 100$, or as a quadrupole mass filter. Behind the quadrupole is a hexapole collision cell for molecular break-up studies or gas-phase studies followed by an orthogonal Time-of-Flight mass spectrometer (ToF MS2) with a resolution of $m/\delta m = 10,000$ and a mass range from 50 to 50,000 Dalton.

3. Current status and outlook

The described setup is currently under construction. Sn^+ , Ag^+ , In^+ and Cd^+ have already been produced by an electrospray ionization source and were measured with the qToF. We are currently investigating the kinematics of the reaction of these ions with methane in the collision cell of the qToF. These studies will serve as guidance for future experiments with the gas catcher, opening the door to establish the CISE technique for experimental studies of short-lived isotopes.

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References

- [1] J.B. Neumayr, et al., The ion-catcher device for SHIPTRAP, Nucl. Instr. Meth. Phys. Res. B 244 (2006) 489–500.
- [2] C. Droese, et al., The cryogenic gas stopping cell of SHIPTRAP, Nucl. Instr. Meth. Phys. Res. B 338 (2014) 126–138.
- [3] M. Ranjan, et al., Design, construction and cooling system performance of a prototype cryogenic stopping cell for the super-frs at fair, Nucl. Instr. Meth. Phys. Res. A 770 (2015) 87–97.
- [4] M. Wada, et al., Slow RI-beams from projectile fragment separators, Nucl. Instr. Meth. Phys. Res. B 204 (2003) 570–581.
- [5] A.A. Kwiatkowski, et al., Precision test of the isobaric multiplet mass equation for the $A=32$, $T=2$ quintet, Phys. Rev. C 80 (2009) 051302.
- [6] A. Shayesteh, et al., Reactions of atomic cations with methane: Gas phase room-temperature kinetics and periodicities in reactivity, J. Phys. Chem. A 113 (2009) 5602–5611.
- [7] J.F. Ziegler, et al., SRIM-the stopping and range of ions in matter (2010), Nucl. Instr. Meth. Phys. Res. B 268 (2010) 1818–1823.