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Purification of Radioactive Ion Beams by Photodetachment in a RF Quadrupole Ion Beam Cooler

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Abstract. A highly efficient method for suppressing isobar contaminants in negative radioactive ion beams by photodetachment is demonstrated. A laser beam having the appropriate photon energy is used to selectively neutralize the contaminants. The efficiency of photodetachment can be substantially improved when the laser-ion interaction takes place inside a radio frequency quadrupole ion cooler. In off-line experiments with ion beams of stable isotopes, more than 99.9% suppression of Co⁻, S⁻, and O⁻ ions has been demonstrated while under the identical conditions only 22% reduction in Ni⁻ and no reduction in Cl⁻ and F⁻ ions were observed. This technique is being developed for on-line purification of a number of interesting radioactive beams, such as ⁵⁶Ni, ^{17,18}F, and ^{33,36}Cl.

Keywords: Photodetachment, isobar suppression, buffer gas cooling; RF quadrupole ion guide.

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INTRODUCTION

The Holifield Radioactive Ion Beam Facility (HRIBF) at the Oak Ridge National Laboratory is an Isotope Separator On-Line (ISOL) facility, providing high-quality radioactive ion beams (RIBs) for studies of exotic nuclei and nuclear astrophysics research. The intensity and purity of the RIBs are of crucial importance. Unfortunately, the RIBs produced using the ISOL technique are often mixtures of the radioactive isotope of interest and isobaric contaminants that cannot be removed effectively by magnetic separation. Many experiments, including some of the most interesting and important, can be compromised by even a modest fraction of impurity ions in the beam. Consequently, development of additional beam purification techniques is necessary. The HRIBF tandem post-accelerator requires negatively charged ions as input. Selective removal of unwanted negative ion species by photodetachment has been suggested for applications in accelerator mass spectrometry [1,2], as well as for purification of RIBs at HRIBF [3]. In this method, a laser beam having the appropriate photon energy is used to selectively neutralize the contaminant if the electron affinity of the contaminant is lower than the electron affinity of the desired radioactive ions. However, in the pioneering work by Berkovits et al. [1,2], the overall degree of isobar suppression reported was too small to be useful.

We have proposed a novel scheme [4,5] that has been demonstrated to be highly efficient and practically suited for use at HRIBF as well as other ISOL and accelerator

mass spectrometry (AMS) facilities. In this new scheme, laser-ion interaction is made inside a radio frequency quadrupole (RFQ) ion beam cooler where the ion residence time can be on the order of milliseconds. As a result, the photodetachment efficiency can be dramatically increased. In the initial proof-of-principle experiment [4], 95% suppression of $^{59}\text{Co}^-$ ions was obtained while under identical conditions only 10% of $^{58}\text{Ni}^-$ ions were neutralized. In this paper, we report the latest experimental results on selective suppression of S, O, and Co negative ions.

PHOTODETACHMENT TECHNIQUE

The photodetachment process



takes place when the photon energy, $h\nu$, is larger than the threshold energy, or the electron affinity (EA) of the atom A , for detaching the electron from the negative ion. Photodetachment, then, can be used to neutralize and hence remove unwanted negative ions from beams. This principle can be applied to isobar suppression if the electron affinity of the isobaric contaminant, EA_1 , is lower than the electron affinity of the desired radioactive ions, EA_2 , i.e., $EA_1 < EA_2$. In this case, it is possible to selectively neutralize the isobar contaminants by photodetachment with photons of energy $EA_1 < h\nu < EA_2$.

For a given laser power and photon energy, the fraction of negative ions not neutralized by the laser radiation, assuming complete overlap of the ion and laser beams, is given by

$$\frac{n}{n_0} = \exp(-t\sigma\phi) \quad (2)$$

where n_0 is the initial number of negative ions before interaction with the laser radiation, n is number of ions not neutralized by the laser, ϕ is the laser flux (photons $\text{cm}^{-2}\text{s}^{-1}$), σ is the photodetachment cross-section (cm^2), and t is the laser-ion interaction time (s). The cross-section for photodetachment is typically on the order of 10^{-17} cm^2 . To obtain high photodetachment efficiency and thus a high degree of isobar suppression, very large photon fluxes or long interaction times are required.

Our initial motivation for exploring the photodetachment technique is the desire to purify the ^{56}Ni beam at HRIBF, which is dominated by an order of magnitude more abundant ^{56}Co contaminant. According to Eq. 2, for 99% photodetachment of $^{56}\text{Co}^-$ by 1064 nm (1.1653 eV) laser radiation with a cross-section of $6 \times 10^{-18} \text{ cm}^2$ [2], the product of laser power density and interaction time is $\sim 0.14 \text{ (Ws/cm}^2\text{)}$, e.g., a 100 W/cm^2 laser power density for a 1.4 ms interaction time. However, for 20 keV $^{56}\text{Co}^-$ ions, 1.4 ms implies a spatial laser-ion overlap of over 360 m.

We proposed a new technique of photodetachment in a RFQ ion cooler [4,5], which can substantially improve the efficiency of photodetachment processes, and consequently the degree of isobar suppression. As described in the following Sections, this new technique promises near 100% suppression of isobar contaminants in

negative ion beams in a setup compact enough to easily implement at existing accelerator facilities.

DESCRIPTION OF THE RF QUADRUPOLE ION COOLER

A RFQ ion cooler is a gas-filled RFQ ion guide, which has been widely used as an ion beam cooler and buncher for use with radioactive ion beams [6-12]. In such an ion cooler, ions lose energy rapidly in collisions with the buffer gas molecules. The mean kinetic energy of the ions can be reduced to approximately the thermal energy of the buffer gas and the ion trajectories can be confined to a small region near the longitudinal axis of the device. Once thermalized, the ions move at low velocity through the RFQ under the influence of a small longitudinal electrostatic field gradient. Total transit times can be more than milliseconds. This combination of small transverse ion beam dimension and extended ion transit time provides ideal conditions for photodetachment.

A RFQ ion cooler for use with negative ions has been developed at HRIBF [13, 14]. A schematic view of the negative ion cooler is shown in Fig. 1. The quadrupole consists of four parallel cylindrical rods of 8-mm diameter and 40-cm length, equally spaced with an inscribed circle of radius $R_0 = 3.5$ mm. The quadrupole rod structure is mounted inside a Cu cylindrical enclosure (length: 40 cm, inner diameter: 30 mm) with a 3-mm diameter ion beam entrance aperture and a 2-mm diameter exit aperture. Buffer gas is introduced into the quadrupole assembly through an orifice located near the middle of the Cu cylinder. Negative ions are very fragile. Electron detachment (loss of the negative ion) can take place in collisions with the buffer gas if the ion energies are too high relative to the electron affinity of the ion species. In order to reduce losses due to collisional electron detachment, the ions are decelerated to less than 40 eV before being injected into the quadrupole. When the ions reach the exit aperture they will be re-accelerated to high energies.

After being cooled to near thermal energies, the ions move randomly in the axial direction and it is necessary to provide a longitudinal field to guide them out. Four DC electrodes are placed between the quadrupole rods (Fig. 2a). The DC electrodes are tapered at a very small angle with respect to the quadrupole axis, such that the distance between the electrodes changes from 10 mm at the entrance to 11 mm at the exit plane of the quadrupole. When the DC electrodes are negatively biased relative to the quadrupole electrodes, a weak longitudinal field along the axis of the quadrupole is created that gently pushes the negative ions toward the exit aperture. According to simulations conducted using the finite element code ANSYS [15], the axial potential difference across the whole quadrupole length is about 1 V per -20 V bias on the DC electrodes and the average longitudinal field increases linearly with the potential difference between the DC and RF electrodes as shown in Fig. 2b.

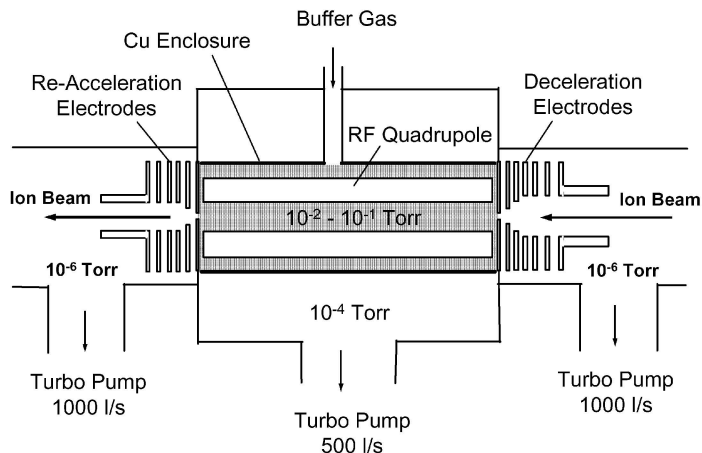


FIGURE 1. Schematic view of the RFQ ion cooler coupled with deceleration and re-acceleration electrodes. Ions enter the RFQ from the right through the 3 mm entrance aperture and exit through the 2 mm exit aperture on the left.

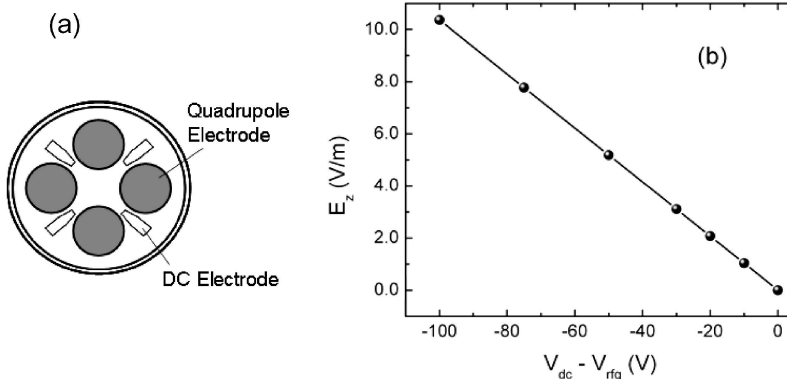


FIGURE 2. (a) Schematic view of the RF and DC electrode assembly. (b) Average axial longitudinal field created by the tapered DC electrodes as a function of the potential difference between the DC and the RF electrodes, as calculated using ANSYS.

The cooler typically operates at RF frequencies of around 2.75 MHz and RF peak voltages up to 500 V, with He as the buffer gas. High transmission for negative radioactive ion beams is required for the HRIBF research program. There are inherent difficulties in cooling and transporting negative ions through a gas-filled RF quadrupole ion guide because negative ions are much more fragile than their positive-ion counterparts. Significant improvements in its performance have been reported [14]. Table 1 presents the cooler transmission efficiencies measured for a number of negative ions.

TABLE 1. Ion Cooler Transmission Efficiency

Negative Ion	Mass (amu)	EA (eV)	Transmission (%)
O	16	1.4611	24 ± 4
F	19	3.3993	38 ± 8
S	32	2.0771	45 ± 5
Cl	35	3.6173	51 ± 8
Ni	58	1.1561	52 ± 12
Co	59	0.6611	43 ± 13
Cu	64	1.2281	52 ± 11

Ion Residence Time in the RFQ Ion Cooler

A Monte Carlo model has been developed to simulate the transport of ions through the RFQ ion cooler [13]. Simulations show that the ion residence time inside the cooler is affected by ion injection energy, buffer gas pressure inside the quadrupole and the longitudinal field strength created by the tapered DC electrodes. Fig. 3 shows the simulated average transit times for $^{56}\text{Co}^-$ ions at different He buffer gas pressure and different longitudinal fields, with typical ion injection energy 17-40 eV. As expected, the time that an ion may spend inside the cooler increases with increasing buffer gas pressure and decreasing longitudinal drift field. The cooler typically operates with He pressures ranging from about 3 to 6 Pa and the tapered DC electrodes biased at 0-50 V more negative than the RF rods. According to Fig. 2 and 3, ion residence times of ≥ 1 ms could be easily obtained.

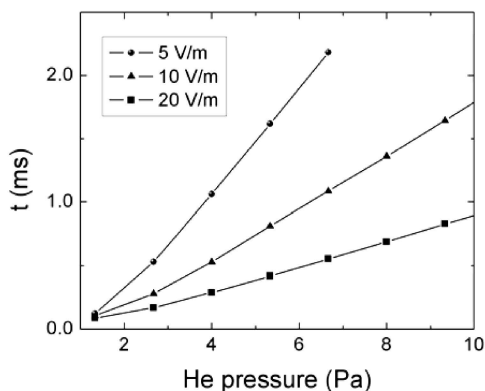


FIGURE 3. Calculated average residence time of $^{56}\text{Co}^-$ negative ions in the RFQ ion cooler as a function of He buffer gas pressure. The $^{56}\text{Co}^-$ ions have initial energies of 17-40 eV.

EXPERIMENT

The feasibility of photodetachment in a RFQ ion cooler has been investigated using negative ions of stable isotopes at the off-line Ion Source Test Facility I (ISTF-1) of

HRIBF. Our goal is to study collisional cooling and photodetachment processes in the ion cooler, optimize the cooler design and the optical system for laser beam transportation and coupling, and eventually implement this technique at the HRIBF for purifying isobaric contaminants in RIBs.

There are a number of interesting radioactive beams, such as $^{17,18}\text{F}$, ^{33}Cl , and ^{56}Ni , needed for studies in nuclear structure and nuclear astrophysics at HRIBF. However, these beams are often dominated by an isobaric contaminant, $^{17,18}\text{O}^-$, $^{33}\text{S}^-$, and $^{56}\text{Co}^-$, respectively, resulting in low-purity radioactive beams not desired for experiments. The electron affinities of these negative ions are F (3.3993 eV), O (1.4611 eV), Cl (3.6173 eV), S (2.0771 eV), Ni (1.1561 eV) and Co (0.6611 eV). In each case, a laser beam having the appropriate photon energy can be used to selectively neutralize the isobar contaminant. Our effort has been focused on developing the technique for these RIBs.

A detailed description of the experimental apparatus has been given previously [4,5]. Briefly, negative ions were produced with a Cs-sputter negative ion source. After acceleration to about 5 keV, the negative ions were separated in mass by a 90° dipole magnetic and the mass-selected ions were sent into the RFQ ion cooler. The ion current before the cooler was measured with a Faraday cup. Ions emerging from the cooler were deflected by an electrostatic deflector to an off-axis Faraday cup detector. For photodetachment, a laser beam was sent into the cooler through the 2 mm exit aperture, traveling collinearly with but in the opposite direction of the ion beam propagation. The laser beam intensity was measured before entering the experimental chamber and after passing through the ion cooler and emerging from a vacuum window at the mass-separation magnet. The interaction of the laser with the negative ions was studied by monitoring the changes in the ion current obtained at the off-axis Faraday cup when the laser beam was on.

RESULTS

Photodetachment in the RFQ Ion Cooler

A pulsed Nd:YLF laser with frequency-doubled output at 527 nm was used for photodetachment of S and O. The photon energy, $h\nu = 2.3526$ eV, is larger than the electron affinity of S^- and O^- but smaller than that of F^- and Cl^- . Therefore, S^- and O^- ions were expected to be strongly neutralized by the 2.3526 eV photons with Cl^- and F^- unaffected. The laser provided short photon pulses of 10 – 30 ns at variable rates of 1 Hz – 10 kHz with maximum average power of about 3 W at 3 kHz.

A set of typical data are illustrated in Fig. 4. Fig. 4a shows the fraction of $^{32}\text{S}^-$ ions not neutralized by the laser beam versus laser power obtained at different He gas pressures, plotted in logarithmic scale. The cooler was operated at RF frequency of 2.75 MHz with peak RF amplitude of $V_{\text{rf}} \sim 150$ V. The $^{32}\text{S}^-$ ions were decelerated to about 20 eV when entering the quadrupole and the bias voltage on the tapered DC electrodes was optimized for ion transmission. The fraction of the surviving ions was determined as the ratio of the ion current measured at the off-axis Faraday cup when

the laser was on to that with the laser off. It can be seen that the ratios, n/n_0 , became saturated at gas pressures < 5.7 Pa. One possible explanation is that at relatively low He pressures, the ion trajectories in the radial direction were not sufficiently damped and the spatial distribution of the ions was larger than the laser beam size. When the He pressure was increased to 5.7 Pa, the surviving ion fraction decreased exponentially with increasing laser power, as given by Eq. 2. A factor of 1000 reduction of the $^{32}\text{S}^-$ ions was obtained with only about 1 W laser power. This may indicate that the ions were sufficiently cooled and their spatial distribution in the radial direction was reduced to be comparable with or smaller than the laser beam size.

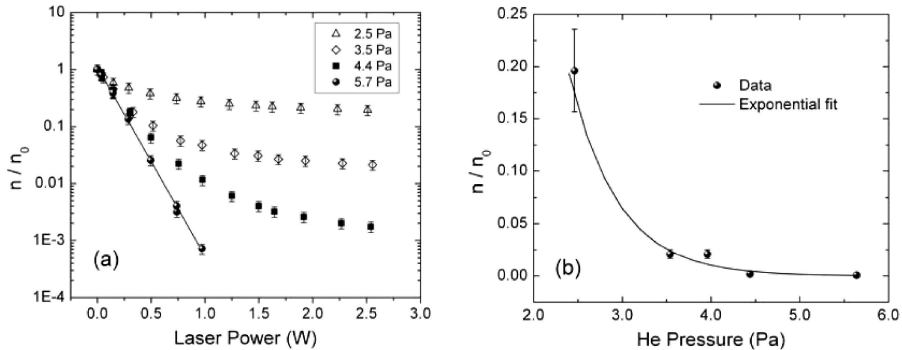


FIGURE 4. (a) The fraction of surviving $^{32}\text{S}^-$ ions versus laser power measured at a different He pressures. The efficiency of photodetachment was saturated at lower He pressures. (b) The fraction of surviving ^{32}S negative ions versus He buffer gas pressure at a fixed laser power of 2.5 W. The data fit reasonably well to an exponential decay function.

For a fixed laser power, the photodetachment efficiency also increased exponentially with increasing buffer gas pressure, as shown in Fig. 4b. This implies that the ion residence time in the cooler increased linearly with increasing buffer gas pressure, in agreement with the simulation results (Fig. 3).

The laser pulse rate was variable from 1 Hz to 15 kHz. It was found that more S^- ions were depleted at higher laser repetition rates, even though the average laser power was about the same. Fig. 5 shows the ratio of n/n_0 as a function of the laser repetition rate, obtained at laser power of 2.5 W and two different He pressures. The DC electrodes were biased at about -40 V relative to the RF electrodes, which corresponds to a longitudinal field of $E_z \sim 4$ V/m. In both cases, the fraction of surviving $^{32}\text{S}^-$ ions decreased with increasing laser repetition rate but reached a saturation level above about 10 kHz. The results indicate that at low pulse repetition rates, e.g., less than 3 kHz, the laser pulse duty cycle was too low that a substantial fraction of the ions could pass the ion cooler without interaction with the photons. Increasing the pulse repetition rate increased the laser pulse duty cycle and thus the probability for more ions to interact with the photons, resulting in higher photodetachment efficiency. Above 10 kHz, most of the ions encountered at least one laser pulse during their passes through the cooler. Thus, the overall photodetachment efficiency was saturated.

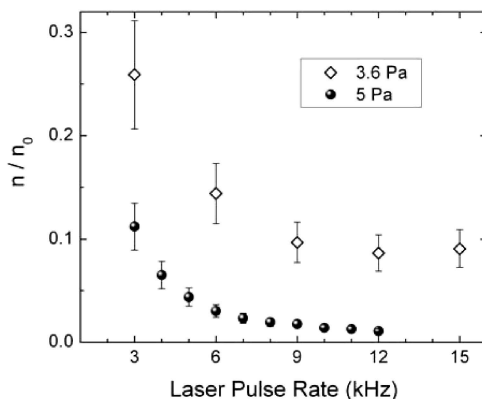


FIGURE 5. The fraction of surviving $^{32}\text{S}^-$ ions as a function of the laser pulse rate, obtained at He pressure of 3.6 Pa and 5 Pa. The laser power was fixed at 2.5 W.

The efficiency of photodetachment was also affected by the longitudinal field created by the tapered DC electrodes. Fig. 6 shows the fraction of $^{32}\text{S}^-$ not neutralized as a function of the laser power, obtained at two different bias potentials on the tapered DC electrodes, $\Delta V = V_{\text{dc}} - V_{\text{rfq}}$. A larger negative value of ΔV corresponds to a larger longitudinal field that pushes the ions toward exit (Fig. 2b), and in turn a shorter transit time for the ions. Therefore, smaller photodetachment efficiency is expected for a larger negative bias potential on the DC electrodes. This was experimentally observed. The measurements were made at about 6 Pa He pressure in the quadrupole. Again, the data could fit well to an exponential decay and 99.9% of the $^{32}\text{S}^-$ ions were depleted with only about 1 W laser when $\Delta V = -10$ V.

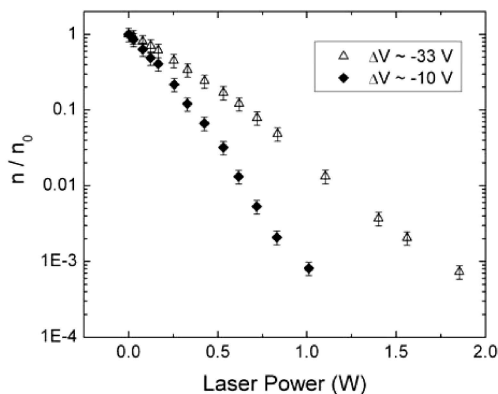


FIGURE 6. The fraction of surviving $^{32}\text{S}^-$ ions as a function of laser power, obtained at two DC electrode bias potentials. As expected, larger bias, thus larger longitudinal field, resulted in lower photodetachment efficiency.

The effective interaction time between the laser and $^{32}\text{S}^-$ ions under different cooler operation conditions was estimated by fitting the corresponding photodetachment data, n/n_0 versus laser power, to Eq. 2. A photodetachment cross section of $1.0 \times 10^{-17} \text{ cm}^2$ for S^- [1] was used for the calculations. The preliminary results, calculated using a simple assumption of an effective laser beam size of 2 mm in diameter, are plotted in Fig. 7. These data were obtained at DC electrode bias $\Delta V = -3 \text{ V}$, which corresponded to a very small longitudinal field. The estimated interaction times are between 4 to 8 ms, increasing linearly with increasing He pressure, as expected by simulations (Fig. 3). Further analysis will take into account the photon and ion spatial distributions.

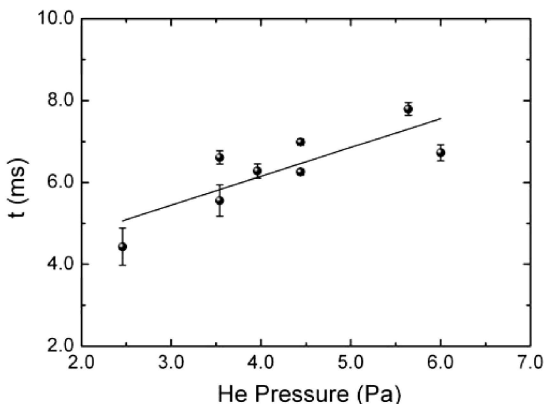


FIGURE 7. The effective laser-ion interaction time for $^{32}\text{S}^-$ ions, calculated by fitting the n/n_0 versus laser power data to Eq. 2. The RFQ ion cooler was operated with $\Delta V = -3 \text{ V}$ bias on the DC electrodes.

Selective Suppression of the (S, Cl) and (O, F) Pairs

Near 100% photodetachment efficiency was obtained at $\sim 6 \text{ Pa}$ He pressure. Fig. 8 shows a temporal recording of the $^{32}\text{S}^-$ and $^{35}\text{Cl}^-$ current measured at the off-axis Faraday cup. When the 527 nm laser beam of 2.5 W was turned on, the $^{32}\text{S}^-$ current dropped immediately from about 6.3 nA to 2 pA, which was the baseline of our data acquisition system. This corresponded to a photodetachment efficiency of about 99.97%, or a factor of 3000 depletion of the $^{32}\text{S}^-$ ions. When the laser beam was blocked, the ion current quickly returned to the initial values. The photo-depletion process was fast and reproducible as shown by the data. Under the same conditions, no depletion of $^{35}\text{Cl}^-$ beams was observed. The photodetachment efficiency was also measured without the use of the RFQ ion cooler. In this case, the laser beam interacted with 5 keV $^{32}\text{S}^-$ beam and only about 1% $^{32}\text{S}^-$ ions were depleted by 2.5 W laser power.

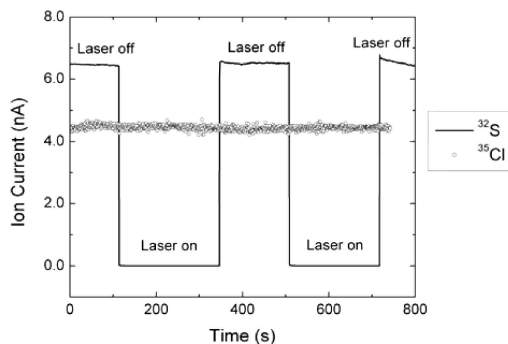


FIGURE 8. Measured intensities of $^{32}\text{S}^-$ and $^{35}\text{Cl}^-$ ion beams accelerated from the RFQ ion cooler. 99.97% of $^{32}\text{S}^-$ ions were neutralized by 2.5 W laser ($\lambda = 527$ nm), while the $^{35}\text{Cl}^-$ were not affected. He pressure 6 Pa, $f = 2.76$ MHz, $V_{rf} \sim 200$ V, DC electrode bias $\Delta V = -10$ V.

Similar results were obtained for O^- and F^- ion pairs. As shown in Fig. 9, 99.9% of the $^{16}\text{O}^-$ ion beam was depleted by ~ 2.5 W laser power (Fig. 9a), while no neutralization of the $^{19}\text{F}^-$ beams was observed (Fig. 9b).

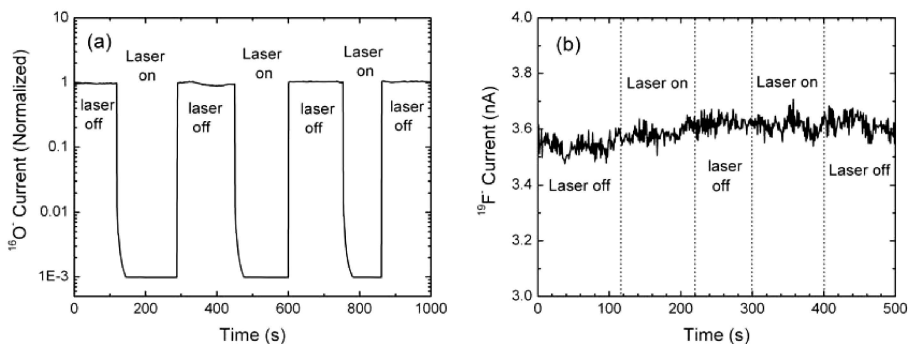


FIGURE 9. (a) $^{16}\text{O}^-$ and (b) $^{19}\text{F}^-$ currents measured at the off-axis Faraday cup. The $^{16}\text{O}^-$ current shown was normalized to the ion current when the laser beam was off. 99.9% of the $^{16}\text{O}^-$ ions were neutralized by 2.5 W laser ($\lambda = 527$ nm). No reduction in the $^{19}\text{F}^-$ beam was observed. He pressure ~ 6 Pa, $f = 2.76$ MHz, $V_{rf} \sim 75$ V, DC electrodes bias $\Delta V = -5$ V.

Photodetachment of the (Co, Ni) Pair

A continuous wave (CW) Nd:YAG laser at 1064 nm ($h\nu = 1.1653$ eV) was used to selectively remove the Co^- . The photon energy was well above the electron affinity of Co^- (0.6611 eV), but also slightly larger than the electron affinity of Ni^- (1.1561 eV). Therefore, some neutralization of Ni^- ions was expected. Figure 10 shows the $^{59}\text{Co}^-$ and $^{58}\text{Ni}^-$ currents measured by the off-axis Faraday cup when the RFQ ion cooler was operated at a He pressure of ~ 10 Pa and the laser beam was turned on and off. About 99.9% photodetachment efficiency for $^{59}\text{Co}^-$ was achieved with about 4 W laser power measured at the output of the laser while under the same conditions, about 22%

depletion of Ni^- was observed. When the ion cooler was off and the ions were moving at about 5 keV energy, only about 3% depletion in the $^{59}\text{Co}^-$ current, and no change in the $^{58}\text{Ni}^-$ current, was observed with 4 W laser radiation. .

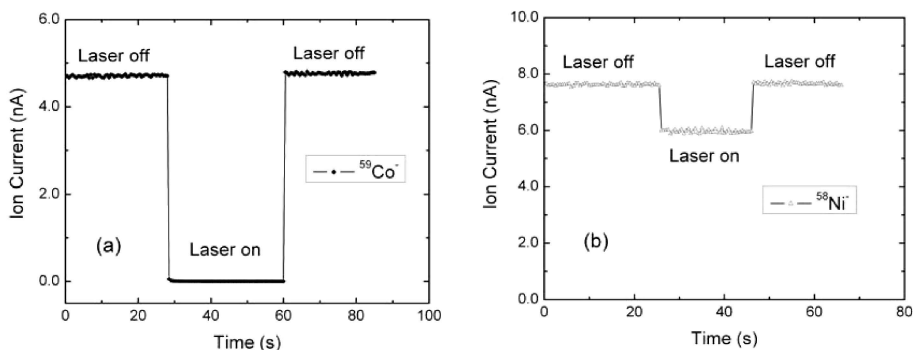


FIGURE 10. Measured intensities of (a) $^{59}\text{Co}^-$ and (b) $^{58}\text{Ni}^-$ negative ion beams through the RFQ cooler at a He pressure of ~ 10 Pa. 99.9% of the $^{59}\text{Co}^-$ and 22% of the $^{58}\text{Ni}^-$ ions were neutralized with a CW laser beam of 4 W. RF = 2.75 MHz, $V_{rf} \sim 300$ V, and DC electrode bias $\Delta V = -40$ V.

CONCLUSION

The feasibility of efficient isobar suppression by photodetachment in a gas-filled RFQ ion guide has been experimentally demonstrated with ion beams of stable isotopes. Laser beams having the appropriate photon energy were used to selectively neutralize the contaminants if the electron affinity of the contaminants was lower than the electron affinity of the desired radioactive ions. Simulation and experimental results showed that the photodetachment efficiency could be dramatically increased when the laser-ion interaction took place inside a RF quadrupole ion beam cooler where the laser-ion interaction time was on the order of milliseconds. For the selected negative ion pairs of (S, Cl) and (O, F), 99.97% of $^{32}\text{S}^-$ and 99.9% of $^{16}\text{O}^-$ ions were depleted with only about 2.5 W average laser radiation of 527 nm photons from a pulsed laser, while the desired Cl^- and F^- ions were not affected. In the case of the (Co, Ni) pair, 99.9% suppression of $^{59}\text{Co}^-$ ions was demonstrated with a CW Nd:YAG laser beam of 4W power at 1064 nm, with only about 22% of $^{58}\text{Ni}^-$ ions suppressed. The depletion of Ni^- ions can be avoided by using photons of energy less than the electron affinity of Ni. The laser power used to achieve nearly 100% photodetachment is readily available from existing commercial lasers and the setup is compact enough to easily implement at an existing accelerator facility. Therefore, this technique is very promising for real applications, such as purifying isobaric contaminants in radioactive ion beams for nuclear research or accelerator mass spectrometry. It is being developed for use at the HRIBF for on-line purification of a number of interesting radioactive beams, such as ^{56}Ni , $^{17,18}\text{F}$, and ^{33}Cl . Applications in accelerator mass spectrometry to suppress the ^{36}S contaminants in ^{36}Cl beams will also be explored.

ACKNOWLEDGMENTS

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