Submillisecond On-Line Mass Separation of Nonvolatile Radioactive Elements: An Application of Charge Exchange and Thermalization Processes of Primary Recoil Ions in Helium

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Transportation of thermalized primary recoil ions from nuclear reactions by helium flow has been investigated as a means of injecting short-lived radioactive nuclides into an on-line isotope separator. Several short-lived radioactive isotopes of highly nonvolatile elements such as B, Sc, Nb, and W have been separated. The efficiency for heavy nuclides with half-lives above 1 ms is between 1 and 10%. The shortest-lived activity identified in an on-line separation is the 182-μs isomeric state in ^{207}Bi.

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Recent progress in studies of exotic nuclides far off beta stability[1-3] is largely due to the rapid development of on-line mass analyzing devices. On-line isotope separators equipped with fast and efficient ion sources have played a central role in studies and discoveries of several new nuclear properties of highly unstable nuclei.[2-4] For these ion sources, the ionization of almost all elements has become possible.[5] However, the problem is still the long release time of most nonvolatile elements out from the ion source. In this Letter we wish to report the first results of the ultrafast $\gtrsim 100$ μs) on-line mass separation of highly refractory elements using the recently constructed ion-guide isotope-separator on-line (IGISOL) system.[6,7] The principle of operation is based on a thermalization of primary recoil ions in helium and on their subsequent transfer by a helium flow through a differential pumping system into the acceleration stage of a mass separator. With this approach, separation times of several orders of magnitude shorter than those typical of the ion-source-based systems are achieved.

Energetic residual atoms produced in the nuclear reactions are in an ionized state, with their charge state proportional to the velocity. During thermalization, these fast-moving ions change their charge state continuously via charge-exchange processes with the atoms of the medium.[8] In (pure) helium the slow-velocity recoils will keep their low charge states because of the high ionization potential of helium. In the case of +2-charged recoil ions, impurity molecules, such as O_2 and N_2, can cause notable charge transfer $+2 \rightarrow +1$ at thermal energies, but are unable to neutralize singly charged ions because of the endoergicity of those reactions.[9] The presence of an accelerator beam in the stopping gas creates a weakly ionized plasma[10] and complicates the situation; additional charge-transfer and three-body recombination processes in gas become possible. The role of these processes is at present not well understood. However, our experience with thermalized recoils produced in light-ion-induced compound nuclear reactions shows that a large fraction of the recoils possess a charge state +1 over a period of time long enough to be transported by a gas flow out of the thermalizing volume. This finding makes the described principle an attractive choice as an injector for an on-line mass separation of short-lived radioactive nuclides.

Ions of short-lived radioactive atoms were produced via p-, d-, ^{3}He-, and alpha-induced reactions in solid targets of a few milligrams per square centimeter thickness. Reaction recoils are stopped in helium buffer gas at a pressure of 10 kPa within an effective volume of about 1 cm^3. Subsequently, the thermalized ions are swept along with a 30-cm^3/s (STP) flow through a 1.2-mm-diam exit hole into an adjacent vacuum chamber. The positive ions are directed by an electric field over a distance of 10 mm through a 1.5-mm-diam skimmer hole into the acceleration chamber of the mass separator, while most of the helium is removed from the vacuum chamber by means of a high-speed (2000 m^3/h) Roots blower. The combination of a small thermalizing volume and a high flow rate results in a short delay time, a condition found necessary for efficient transportation of recoils out of the target chamber. The mass separator coupled to the ion transportation system is of Scandinavian type with a 55° analyzing magnet. The mass resolving power for the setup has been measured to be ≤ 400 at full width at half maximum and 200 at full width at tenth maximum, being of the same order as typically achieved with the on-line ion-source-based systems. The technical details of the device are given in Refs. 6 and 7

TABLE I. Properties of some radioactive recoil nuclides used in this work. E_R is the initial recoil energy, determined on the basis of conservation of linear momentum excluding the effect of the breakup of the assumed compound nucleus. V is the initial recoil velocity expressed relative to the first Bohr orbital velocity for hydrogen, $v_0 = 2.19 \times 10^6$ m/s. $\bar{q}$ is the calculated average charge state of a recoil ion in the target corresponding to energy E_R (see Ref. 12). The half-life, $t_{1/2}$, and the spin-parity, I^π, represent the nuclear properties of the ions. The last column gives the yield of the mass-separated ions. The beam energies were $E_d = 5$ MeV, $E_\alpha = 20$ MeV, and $E_p = 20$ MeV.

Nuclide	Reaction	E_R/nucleon (keV)	V/v_0	$\bar{q}$	$t_{1/2}$ (ms)	I^π	Yield ($1/\mu$A s)
^{12}B	^{11}B(d,p)	59	1.54	2.0	20.2	$\frac{3}{2}^-$	4×10^3
^{43}Sc^m	^{40}Ca(α,p)	41	1.29	4.0	0.435	$\frac{3}{2}^+$	1.6×10^3
^{90}Nb^m	^{90}Zr(p,n)	2.4	0.31	≥ 1	6.2	1^+	1.4×10^3
^{180}W^m	^{181}Ta($p,2n$)	0.6	0.15	< 1	5.5	8^-	0.8×10^3
^{207}Bi^m	^{nat}Pb(p,x)	0.5	0.14	< 1	0.182	$\frac{21}{2}^+$	0.6×10^2

and in a forthcoming publication.[11]

Out of several radioactive ions investigated, a few covering a wide range of nuclear, atomic, and chemical properties are collected in Table I. The yields of singly charged mass-separated ions were determined by standard methods of beta- and gamma-ray spectroscopy. The fraction due to multiply charged ions was found to be less than 1%. The yields in Table I correspond to 0.1–10% fractions of all the recoil ions ejected from the target foils. A remarkable finding is that this fraction does not seem to be too sensitive to the degree of volatility, as demonstrated by a good transmission of ions of the most refractory element, tungsten. The average initial degree of ionization due to stripping in the target foil is a few units of charge for the lightest atoms, but is about or even smaller than one for the heavier ions. The observation of a good fraction of singly charged atomic ions can be explained to be due to a high binding energy, 24.6 eV, of electrons in helium.[13] Ions with low velocities and with low charge states cannot transfer sufficient energy to liberate electrons from helium. This results in substantially reduced capture cross sections. Other charge-transfer and recombination processes are not believed to be important because of the low ionization degree of 10^{-6} of helium. The main losses of ions are either due to inadequate stopping distances for recoils, or due to their diffusion into the walls of the target chamber.

The total transport efficiency determined as the ratio of the mass-separated ions to those recoiling out from the target was measured for isomeric activities of various elements produced with a 20-MeV proton beam. The total recoil yields from the target were determined in beam by observing the intensity of the delayed γ rays deexciting the isomeric state between the micropulses of the cyclotron beam and by normalizing the observed intensity to the target thickness corresponding to the average range of recoils. The recoil ranges in the target were calculated with the semiempirical method of Biersack.[14] The overall efficiency for several nuclides as a function of the half-life is given in Fig. 1(a). It can be seen that the transmission efficiency is between 0.1 and 1.0% for half-lives below 0.5 ms and between 1 and 10% for half-lives above 0.5 ms. With the exception of ^{90}Nb^m, this figure is limited to the $A \approx 200$ region in order to avoid an additional mass-dependent factor on the efficiency, i.e., the loss of recoils on the chamber walls due to inadequate stopping distance in the gas. Figure 1(b) shows a γ spectrum from the mass-separated 5.5-ms 8^- isomer of a highly refractory ^{180}W^m. The absolute efficiency decreases with the increasing range, because the maximum usable gas pressure is limited by the present pumping capacity in relation to the high voltage resistivity of the system. If the efficiency is corrected for the range-related losses, it is found that equal transmissions are obtained for a large fraction of elements, independent of their volatility. A high sensitivity for detecting the short-lived and high-spin isomeric activities is demonstrated by the observation of the 182-μs $\frac{21}{2}^+$ isomer in ^{207}Bi. This corresponds to angular momentum transfer of $\geq 10\hbar$, a value rather high for 20-MeV proton-induced reactions.

The falloff value in the efficiency curve in Fig. 1(a) corresponds qualitatively to the residence time deduced from the measured flow rate. The average evacuation time of the 1-cm^3 target chamber in given conditions is about 3 ms, and the transfer time from the beam axis to the exist hole is about 0.5 ms. From this it can be deduced that the lifetime of thermalized ions in the target chamber must be longer than milliseconds. No essential difference in efficiency was found as a function of the beam intensity between 10 nA and

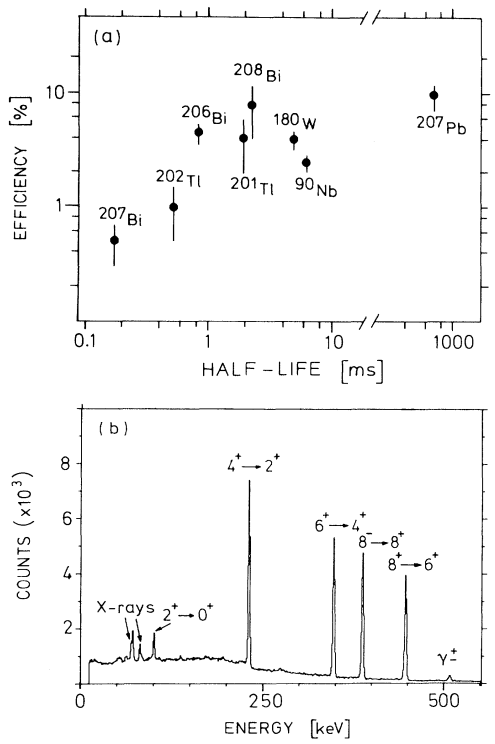

FIG. 1. (a) Overall transmission efficiency for some heavy nuclides in their isomeric states as a function of the half-life. The error bars are due to statistical uncertainties in the measurements. (b) A gamma spectrum from the decay of the mass separated 5.5-ms 8^- isomer of ^{180}W produced via the 20-MeV $p + {}^{nat}$Ta reaction. The total running time was 30 min with the 0.7-μA beam intensity.

1 μA. The effect of the helium pressure on the transmission efficiency could not be studied above the value of ≈ 10 kPa used, because of pumping limitations. The knowledge of this behavior will play a very important role in applications of the technique to heavy-ion reactions where more energetic recoils and hence larger ranges are expected.

In conclusion, we have investigated thermalization of recoil ions and their charge-exchange processes in a high-pressure gas. We have found that a large fraction of thermalized ions remain in the charge state $+1$ over a period of milliseconds. This discovery has allowed us to perform very fast on-line isotope separations of several highly refractory elements. The technique has successfully been used in the studies of nuclear properties of the mirror nuclei ^{47}Cr, ^{51}Fe, and ^{55}Ni and of the $\frac{1}{2}^+$ intruder isomer in ^{203}Bi.[7,15] Further investigations to reach higher helium pressures and hence the possibility of applying this principle to recoils produced in heavy-ion reactions and in fission are in progress.

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