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Abstract. High charge states, up to fully stripped ^{11}C and ^{14}O ion, beams have been produced with the electron cyclotron resonance ion sources (LBNL ECR and AECR-U) at Lawrence Berkeley National Laboratory. The radioactive atoms of ^{11}C and ^{14}O were collected in batch mode with an LN_2 trap and then bled into the ECR ion sources. Ionization efficiency as high as 11% for $^{11}C^{4+}$ was achieved.

INTRODUCTION

BEARS (Berkeley Experiments with Accelerated Radioactive Species) is an initiative to develop a radioactive ion-beam capability at the 88-Inch Cyclotron Facility of Lawrence Berkeley National Laboratory. It involves the production of radioactive isotopes at an existing medical cyclotron on site and a gas-jet transport of the isotopes through a 300 meter capillary to the 88-Inch Cyclotron Facility. The radioactive isotopes are then injected into the electron cyclotron resonance ion sources to be ionized to high charge states and accelerated by the cyclotron for nuclear science experiments. Because of the lower yield of the radioactive isotopes relative to the stable species, efficiencies of target production, transport, ionization and acceleration all play critical roles in the production of radioactive ion beams. In addition to the tests on the target production and transport, developments have been carried out with the ECR ion sources to study and to optimize the ionization efficiencies. The first two radioactive isotopes to be developed are ^{11}C ($t_{1/2}$ = 20.3 min) and ^{14}O ($t_{1/2}$ = 70.6 sec). This article presents and discusses the preliminary results.

ECR ION SOURCES

An electron cyclotron resonance ion source (ECRIS) reliably produces singly to highly charged ion beams with high intensities and high ionization efficiencies. It is the dominant heavy-ion source and has dramatically enhanced the capabilities of many heavy-ion accelerators worldwide.

With its two high charge state ECRISs, the LBNL ECR and the LBNL AECR-U, the 88-Inch Cyclotron has evolved from a light-ion cyclotron into a very versatile accelerator capable of accelerating ions from hydrogen to the heaviest natural element--uranium (1). Other applications of ECRIS are found in atomic physics research, production of radioactive ion beams and industry ion implantation.

The LBNL ECR Ion Source

The LBNL ECR was completed in 1984 and has been reliably operating since then. Its maximum peak magnetic fields on axis are 0.4 and 0.3 Tesla at the injection and extraction regions, respectively. The maximum radial field is 0.3 Tesla at the inner surface of a copper plasma chamber of 45 cm length with a 9 cm inner diameter. There are six large slots between the sextupole magnet bars in the plasma chamber for oven access and radial pumping, as shown in Fig. 1. The conductance of these six slots are calculated to be about 2000 l/s. This ion source operates at 6.4 GHz with a microwave-driven first stage to provide additional cold electrons to the main ECR plasma for the production of highly charged ions. Although the LBNL ECR does not produce ion beams with charge states as high nor intensities as great as the LBNL AECR-U, it continues to provide most of the intermediate charge state ion beams for the cyclotron. The detailed performance of this ion source has been reported in many previous publications (2).

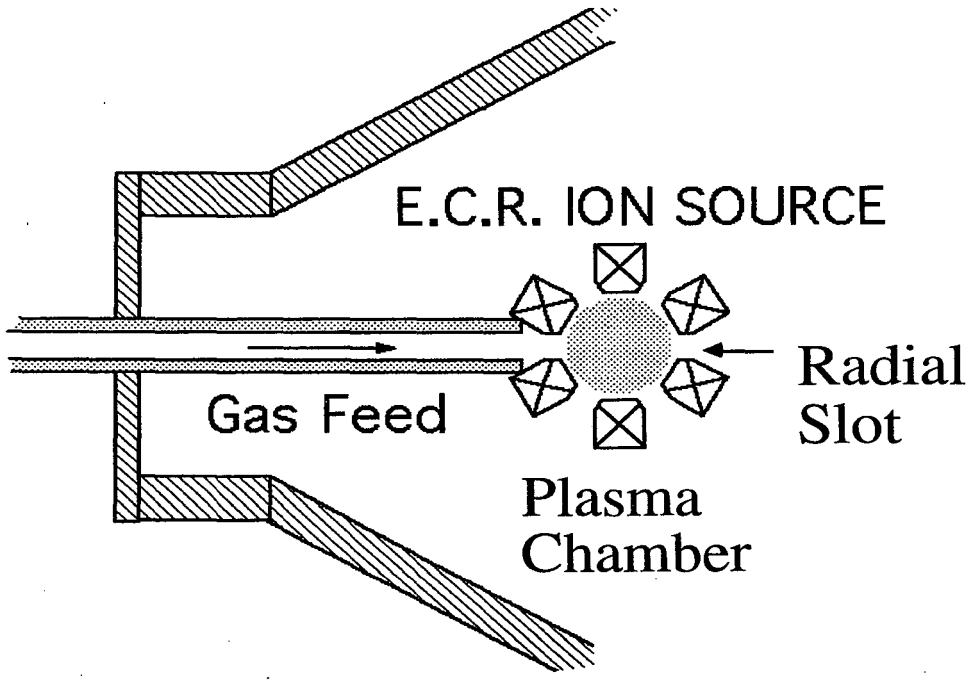

FIGURE 1. The cross section view of the LBNL ECR ion source. The plasma chamber has six large radial slots for pumping and oven access. In the ionization efficiency measurements of ^{11}C and ^{14}O, the activities were fed through one of the radial slots to the plasma chamber.

The LBNL AECR-U Ion Source

The LBNL AECR-U is a higher performance ion source built in 1990 and it was upgraded in 1996 to further enhance its performance. Its maximum peak fields on axis are 1.7 and 1.1 Tesla at the injection and extraction regions, respectively, almost a factor of 4 higher than the LBNL ECR. The maximum radial field at the inner surface of the plasma chamber is 0.85 Tesla. The plasma chamber, made from aluminum for higher yield of secondary cold electrons, is 30 cm in length with an inner diameter of 7.6 cm. Like the LBNL ECR, there are also six slots for pumping and easy oven access in the plasma chamber although the slots are much smaller. The conductance of these six slots are calculated to be about 200 l/s. Shown in Fig. 2 is an elevation view of the LBNL AECR-U ion source. The plasma of the AECR-U is driven by microwaves of two-frequency (14 and 10 GHz) launched through two of the three off-axis waveguides terminated at a bias plate in the injection region. The working gases are bled into the source through one of the waveguides. This ion source is well optimized for the production of highly charged ions (3). It is one of the few ECRISs that have combined all of the recent ECRIS techniques, such as multiple-frequency plasma heating, good plasma chamber surface coating with high yield of secondary cold electrons and high magnetic mirror fields (4). It has produced many record charge states and beam intensities.

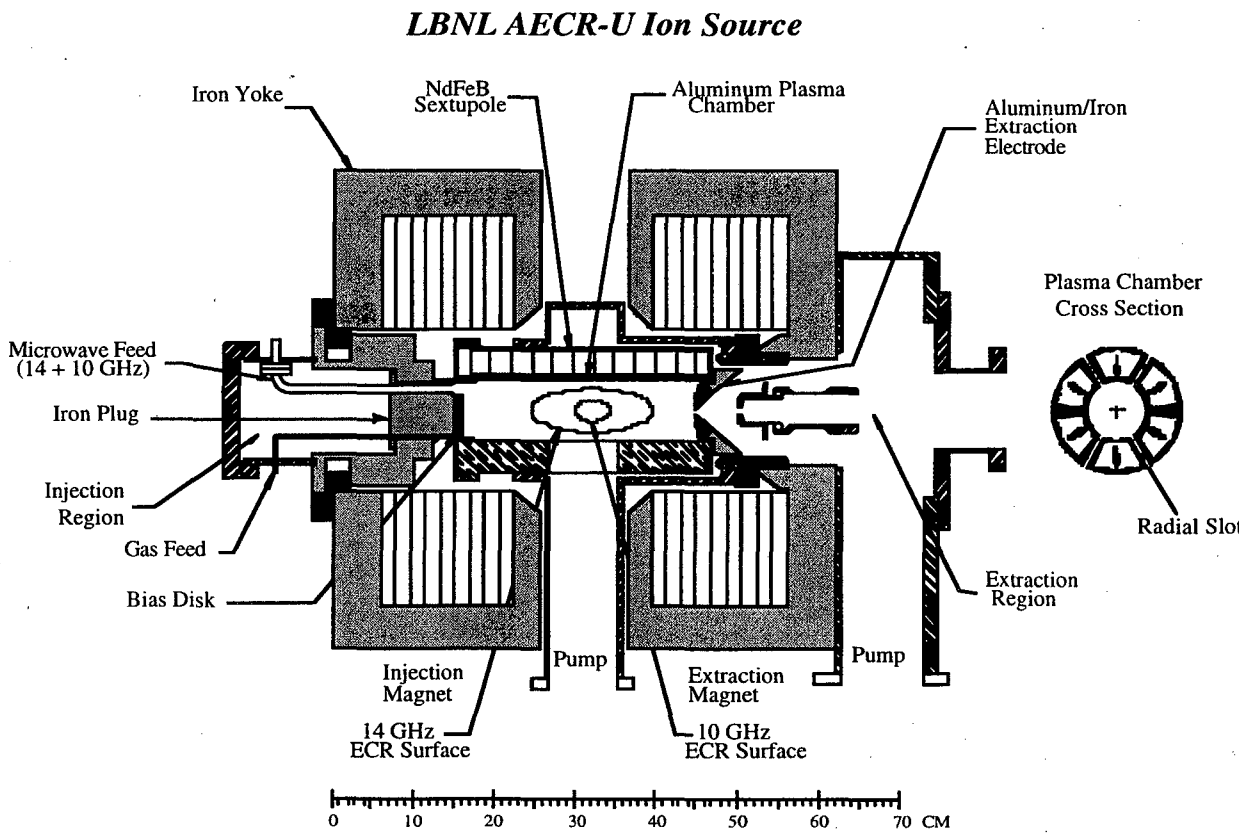

FIGURE 2. An elevation view of the LBNL AECR-U ion source. Only one of the waveguides is shown. The cross section of the plasma chamber is shown on the far right side.

TESTS AND RESULTS

Tests of ionizing the ^{11}C and ^{14}O isotopes were first done with the LBNL ECR ion source. These radioactive isotopes were produced either in a N_2 gas target or a boron nitride (BN) target. A 10 MeV proton beam with intensities up to 5 μA from the 88-Inch Cyclotron was used to irradiate the targets. The initial transport method involved attaching the activity to small aerosol clusters suspended in the carrier gas which is a standard technique which was expected to achieve high transport efficiencies. Unfortunately, it was found that this method failed to transport significant amounts of ^{11}C or ^{14}O. The problem was that most of the activity was in the form of gaseous compounds and did not attach to the aerosol clusters. The fraction of ^{11}C in a chemical form that could be transported was only on the order of 0.1-0.5%. However, this small amount of activity was successfully injected into the LBNL ECR, and an extracted beam of $^{11}C^{1+}$ was identified, although at very low intensities.

The second method, a cryogenic trapping technique, was found to be much more effective for transporting ^{11}C and ^{14}O. Shown in Fig. 3 is the cryogenic trap set-up. The N_2 target/carrier gas was passed through a coil of 3 mm o.d. stainless steel tubing, about 1.5 m long, submerged in liquid nitrogen. About 50% of the total produced activity is successfully trapped. After stopping the gas flow and allowing the remaining nitrogen to be pumped away, the trap was warmed up quickly to 195 °K by an alcohol bath containing dry ice. This temperature increase releases 90% of the trapped ^{11}C (believed to be $^{11}CO_2$) and about 40% of the trapped ^{14}O (believed to be $N^{14}O$ or $N_2{}^{14}O$). The released gas was then radially bled into the LBNL ECR plasma through an adjustable needle valve. Efficient production of high charge state ions from ECRIS requires low pressures, typically of the order of a few $x10^{-7}$ to 10^{-6} Torr, which limit the gas load into the ion source. So care has to be taken to minimize the amount of non-radioactive gas that was trapped and introduced to the ECRIS along with the radioactivity. Otherwise too high a gas flow will greatly reduce the ionization efficiencies of the high charge state ions. By using the dry-ice/alcohol bath, rather than warming the trap to room temperature, the resulting ECR gas load was more controllable and reduced by 90%.

With this batch mode trapping, the ^{11}C activity was measured at the trap before it was released to the ion source. Due to its shorter lifetime, the trapped activity of the ^{14}O in each test run was not measured before releasing to the ion source but the previous measurement of about 50% trapping efficiency of the target yield of ^{14}O was assumed. This assumption of the ^{14}O trapped activity leads to a large uncertainty in determining the ionization efficiency. The ionization efficiency of each ion species is determined by normalizing the measured activity of the particular charge state at a time t back to the trap at t = 0. The release times of the activity from the trap were typically about one minute for the ^{14}O (about one half-life) and a few minutes for ^{11}C (about 0.2 to 0.3 half-life) with trapped radioactivity of about 0.5 to 1 mCi. Although the dry-ice/alcohol bath greatly reduces the gas load, there is still significant amount of stable gases released relative to the ECR ion source mass-handling capability. The short release time and the amount of non-radioactive gases make it unlikely that the ECR source tunings were fully optimized for each charge state as would be the case for constant mass flow in stable beam operation. Nevertheless, with this cryogenic trapping method and the ion source operated at microwave power (6.4 GHz) of 200 to 300 Watts, beams of both ^{11}C and ^{14}O with charge states of up to 5+ were successfully extracted from the LBNL ECR ion source. Listed in Table 1 are the best efficiencies achieved. The ionization efficiency for $^{11}C^{4+}$ was about 0.9% with the LBNL ECR ion source. Similar results were found for 1+, 2+ and 3+ charge states, while the efficiency

for $^{11}C^{5+}$ was only about 0.1%, due to the difficulty in stripping an S-shell electron in the LBNL ECR ion source. Initial measurements of ^{14}O in 3+, 4+ and 5+ charge states indicate efficiencies in the range of about 0.4 - 0.5%.

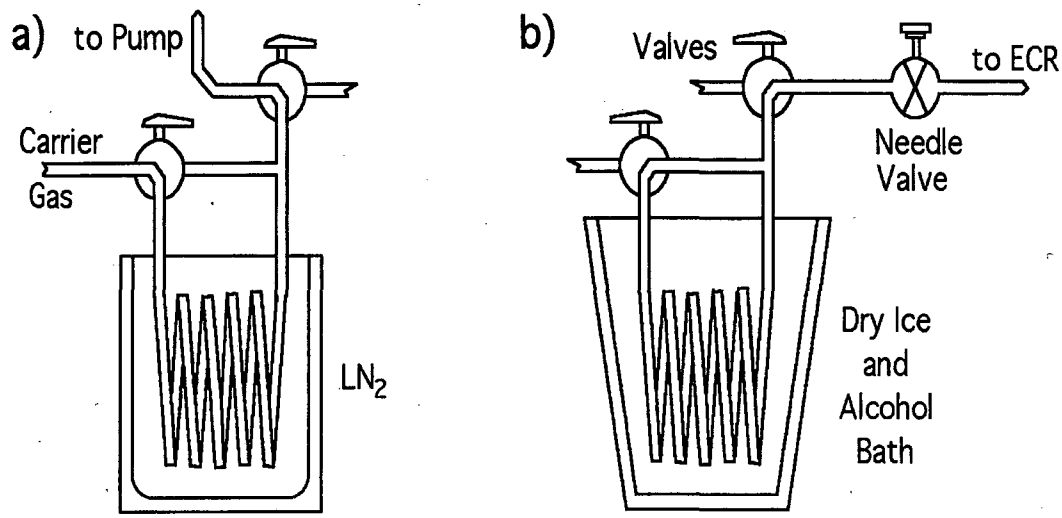

FIGURE 3. Cryogenic trapping system: (a) trapping and (b) release at dry ice temperatures into the ECR ion sources.

Following the tests with the LBNL ECR, the new tests were carried out with the AECR-U to achieve better ionization efficiencies of the high charge state ions. The same cryogenic trapping technique was used to deliver the ^{11}C and ^{14}O activities to the ion source. A different gas injection method was used with the AECR-U. The ^{11}C and ^{14}O isotopes were fed off-axially into the plasma chamber through a 6.3 mm o.d. stainless steel tube terminated at the injection region. Axial injection could achieve a higher trapping of the atoms in the ECR plasma, since the path length of the atoms through the plasma is three times more longer than the path length with the radial injection. In the tests, the AECR-U typically operated with two-frequency plasma heating with total power up to 2.2 kW available from the two klystrons (1.6 kW of 14 GHz and 0.6 kW of 10 GHz) to optimize each charge state with the stable species. As expected, the ionization efficiency for the high charge state ^{11}C and ^{14}O ions are much higher than from the LBNL ECR. High charge states up to fully stripped ^{11}C (2%) and ^{14}O (0.4%) ion beams were extracted from the AECR-U. The AECR-U results are listed in Table 1 in comparison to the results from the LBNL ECR along with the AECR-U ionization efficiencies for stable isotopes of ^{12}C and ^{16}O measured with a calibrated CO leak. The highest efficiency for a single radioactive ion species is 11% for $^{11}C^{4+}$ which is about a factor of 2 lower than the efficiency for $^{12}C^{4+}$. This indicates that there may be still room for further improvement for the ionization of the ^{11}C isotopes.

TABLE 1. Preliminary ionization efficiencies with the ECR and AECR-U ion sources

ION	ECR (%)	AECR-U (%)	AECR-U[a] Stable Species (%)
$^{11}C^{1+}$	1.1		
$^{11}C^{2+}$	0.7		
$^{11}C^{3+}$	0.4	4	
$^{11}C^{4+}$	0.9	11	24
$^{11}C^{5+}$	0.1	4	14
$^{11}C^{6+}$		2	
$^{14}O^{3+}$	0.4		
$^{14}O^{4+}$	0.4		
$^{14}O^{5+}$	0.45		
$^{14}O^{6+}$		3.6	26.7
$^{14}O^{7+}$		1.2	5.6
$^{14}O^{8+}$		0.4	

[a]Stable species are ^{12}C and ^{16}O measured with a calibrated CO leak of flow rate of 9.5 pμA.

DISCUSSIONS AND FUTURE DEVELOPMENT

As shown in Table 1, the ionization efficiencies of the high charge state ions with the AECR-U are much higher than with the LBNL ECR. This is mainly because the AECR-U has a much stronger magnetic field configuration which can support a hotter plasma that is essential to the production of highly charged ions. The other contributions are the off-axial gas introduction, lower chamber conductance and aluminum oxide chamber surface which has a low sticking coefficient. In the LBNL ECR, which has a cooler plasma with presumably lower density, a good portion of the radially injected atoms may just make one radial pass through the plasma and then get pumped away at the opposite large slot or stuck on the copper plasma chamber surface.

In the production of the radioactive ion beams, the source hold-up time can dramatically reduce the ionization efficiency, especially for the short-lived isotopes. The source hold-up time is a function of the source geometry (conductance), plasma chamber surface condition (sticking) and plasma density that determines the ionization length, etc. Figure 4 shows the evolution of the measured activities and the deconvoluted beam rate for $^{14}O^{7+}$ from the AECR-U ion source. The trap valve was opened at time t = 0 and the vertical solid line indicates when the trap valve was closed. The system (transfer line and source) hold-up time for the $^{14}O^{7+}$ is about 20 to 30 seconds. For ^{11}C the system hold-up time is about 8 to 10 minutes which much longer than the ^{12}C hold-up time measured with CO_2 gas on the AECR-U source. We plan to make further tests to resolve the difference.

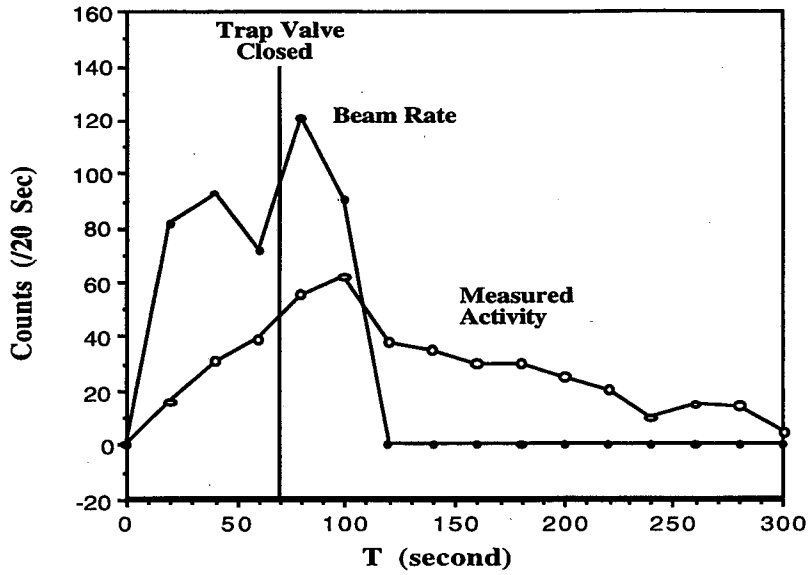

FIGURE 4. Evolution of the measured activity of 2.31 MeV γ-ray and the deconvoluted beam rate of $^{14}O^{7+}$ beam produced with the AECR-U ion source. The solid vertical line indicates the time when the trap was closed. The source hold-up time for the $^{14}O^{7+}$ is about 20 to 30 seconds.

The ionization efficiencies achieved for the high charge state of ^{11}C and ^{14}O ion beams with the AECR-U are comparable to the overall efficiency of the 1+/n+ method using a high efficiency ion source to produce the 1+ radioactive ion beams and then either by accelerating and stripping or injecting the 1+ ions into an ECRIS to produce the desired high charge states (5). If the radiation damage to the permanent magnets is minimized by careful engineering, using an ECRIS to directly and efficiently produce the highly charged radioactive ion beams will be an alternative technique for ISOL type facilities. With this target-ECRIS scheme, the transfer line with various traps can be used to reduce the mass load to the ECRIS. In addition the transfer line can also carry out the selection of certain isobar elements by careful control of the temperatures of the transfer line and traps. The operation of this target-trap-ECRIS system can be cw or batch mode depending on the nature of the desired radioactive species. The detailed of the isobar selection will be addressed in more detail in the future studies.

The first accelerated radioactive ion beam from the 88-Inch Cyclotron was successful carried out recently by transporting the ^{11}C activity in a batch mode from the medical cyclotron in Building 56 to the 88-Inch Cyclotron. With a trap activity of 165 mCi, a $^{11}C^{4+}$ beam of 100 MeV with intensity of 3×10^7 ions/second was extracted from the cyclotron. Figure 5 shows the evolution of the measured activities and the deconvoluted beam rate for the extracted $^{11}C^{4+}$ ions. The system hold-up time is about 10 minutes which is about the same as the measured transfer line and source hold-up time. The total system efficiency of this test is about 0.6% from the trap to the exit of the cyclotron. With further improvements, a total system efficiency of 1 to 1.5% is feasible. So with the ^{11}C production at the medical cyclotron of approximately 1×10^{11} atoms/sec (6), a ^{11}C beam of intensity up to a few $\times 10^8$ ions/sec can be expected for the

completed BEARS system. Similar projections for ^{14}O lead to initial beams of a few x 10^6 ions/sec.

Further tests will be conducted to maximize trap efficiencies and minimize the non-radioactive mass load and transport times. In addition, tests to develop other light-mass proton-rich ion beams, such as ^{13}N, ^{15}O, ^{17}F, ^{18}F and ^{10}C are being considered.

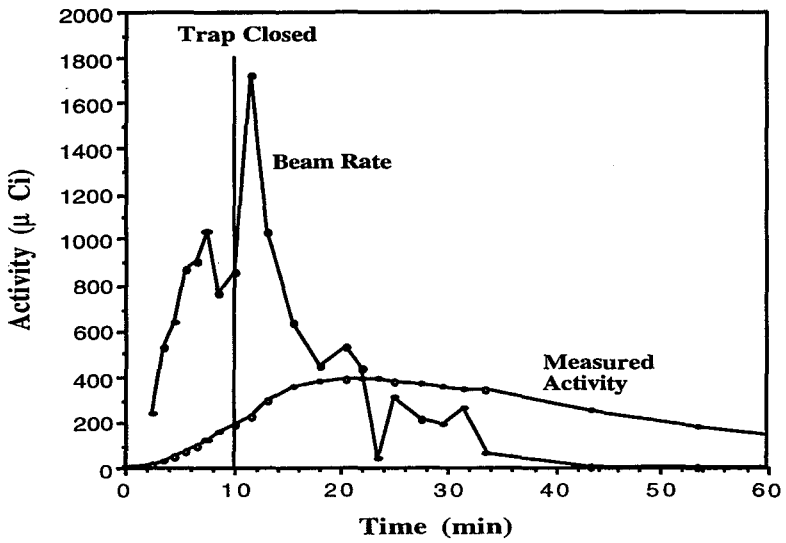

FIGURE 5. Evolution of the measured activity and the deconvoluted beam rate of a 100 MeV $^{11}C^{4+}$ beam extracted from the 88-Inch Cyclotron. The solid vertical line indicates the time when the trap at the AECR-U ion source was closed. The system hold-up time is about 10 minutes.

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