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BEARS: Radioactive Ion Beams at Berkeley

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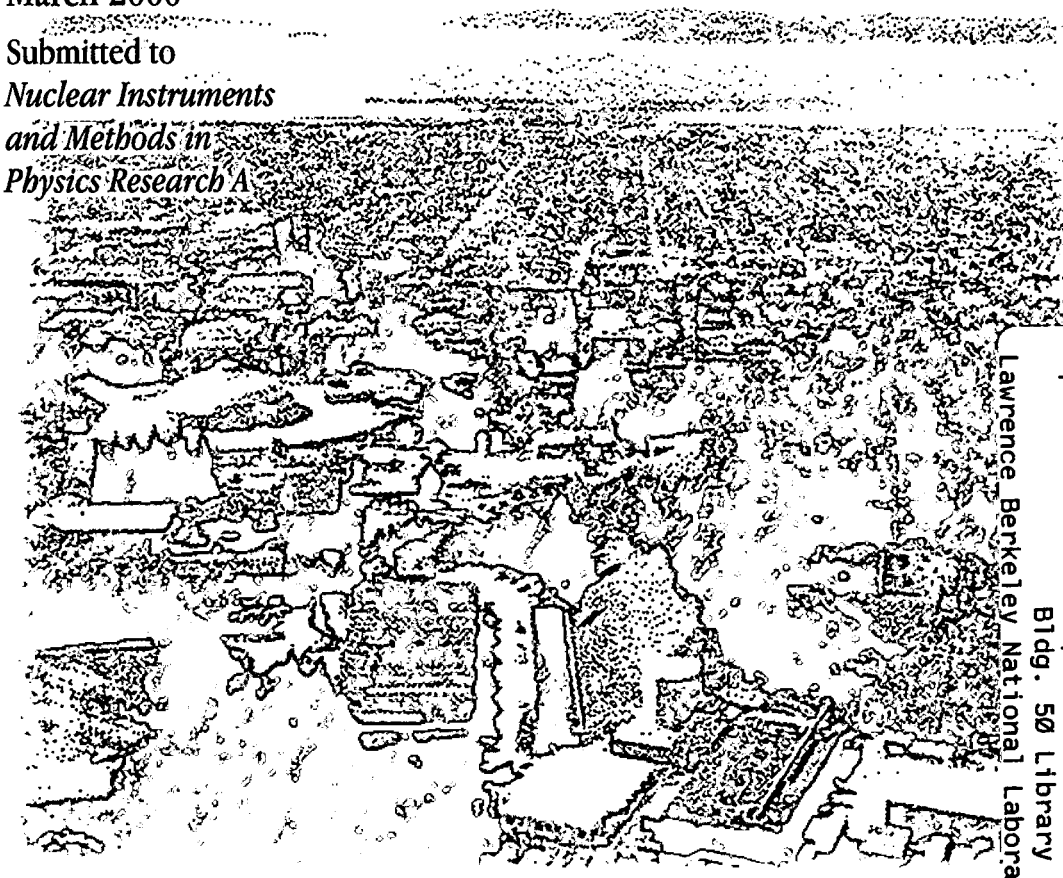

DISCLAIMER

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BEARS: Radioactive Ion Beams at Berkeley

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A light-isotope radioactive ion beam capability has been added to the 88-Inch Cyclotron at Lawrence Berkeley National Laboratory by coupling to the production cyclotron of the Berkeley Isotope Facility. The connection required the development and construction of a 350 m gas transport system between the two accelerators as well as automated cryogenic separation of the produced activity. The first beam developed, ^{11}C, has been successfully accelerated with an on-target intensity of 1 x 10^8 ions/sec at energies of around 10 MeV/u.

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Introduction

This paper describes Berkeley Experiments with Accelerated Radioactive Species (BEARS), a project that adds a light-isotope radioactive ion-beam capability to the 88-Inch Cyclotron at Lawrence Berkeley National Laboratory. The system couples existing equipment for relatively minor cost. After two years of development and construction, BEARS has recently achieved its first steady radioactive beam: carbon-11 with a maximum on-target intensity, at 120 MeV, of 1 x 10^8 ions per second.

The basic BEARS system involves isotope production in a gas target at a low-energy proton cyclotron, transport via a 350-m long gas capillary to the 88-Inch Cyclotron, cryogenic separation of the activity from the target and carrier gases, and injection into the 88-Inch Cyclotron's ion source for ionization and subsequent acceleration.

The production accelerator, normally used to support Positron-Emission Tomography and other medical research, is a 40-microamp, 11-MeV proton cyclotron belonging to the Berkeley Isotope Facility (BIF) [1]. It is capable of producing several light proton-rich isotopes with half-lives long enough to utilize, in particular ^{11}C, ^{13}N, ^{14}O, ^{15}O, ^{17}F, and ^{18}F, all via (p,n) and (p,α) reactions on low-Z targets. Initially, we have focused on the production of ^{11}C ($t_{1/2}$=20 min) and ^{14}O ($t_{1/2}$=71 sec), both produced from a nitrogen gas target. The maximum thick-target production yields are approximately 1 x 10^{11} atoms/sec of ^{11}C and 5 x 10^9 atoms/sec of ^{14}O [2].

The 88-Inch Cyclotron, located about 350 meters from the BIF accelerator at LBNL, is a K=120, sector-focused cyclotron [3], fed by one of two Electron-Cyclotron Resonance (ECR) ion sources. These sources, particularly the upgraded Advanced ECR source (AECR-U) [4] can reliably achieve good ionization efficiencies at high charge states. However, they require vacuums of less than 10^{-6} Torr to operate. Therefore a central technical challenge of BEARS is the coupling of a 350-m long gas transport system to an ECR ion source.

Development Tests:

Prior to construction of an activity transfer line between the two accelerators, tests were carried out entirely at the 88-Inch Cyclotron [5]. A nitrogen-gas cell was bombarded by an 11-MeV proton beam to produce small quantities of ^{11}C and ^{14}O. The target gas flowed continuously from the target, through a plastic capillary of 3-mm inner diameter (i.d.), to an area next to the ion source where it was passed through a cryogenic trap consisting of a stainless-steel coil submerged in liquid nitrogen. A pump at the outlet kept the pressure in the coil well below 1 atm, preventing the condensation of the nitrogen gas, but allowing for the trapping of such gases as CO_2, N_2O, etc. After stopping the flow of target gas and pumping away the residual nitrogen, the liquid nitrogen surrounding the trap was replaced with a dry ice and alcohol bath, raising the temperature enough to release gases such as CO_2 while keeping any water contamination frozen. The gas was slowly passed into the ion source through an adjustable valve, at a rate low enough to prevent overloading of the source. Beams of ^{11}C and ^{14}O with various charge states were extracted from the source, and were measured by the build-up and decay of radioactivity at a Faraday cup located after an analyzing magnet. For each test the ion source was first tuned for the same charge state of a corresponding stable isotope.

The results of these tests were encouraging: about a third of the calculated ^{11}C and ^{14}O production could be successfully trapped and then released, and beams of several different charge states were extracted from the ion sources. A summary of the measured ionization efficiencies is given in Table 1, along with similar results for stable carbon and oxygen, taken with a calibrated CO leak under carefully tuned conditions. The AECR-U was found to have good efficiencies for the radioactive isotopes, with a maximum of 11% for $^{11}C^{4+}$ and 3.6% for $^{14}O^{6+}$. These numbers are lower than the measured stable-isotope efficiencies, possibly because of the relatively high gas load coming from the trap.

On the basis of these results, construction was begun on the transfer line between the two accelerators. In parallel with this effort, development of accelerated ^{11}C beams continued. During the second series of tests, ^{11}C was produced and cryogenically separated at the BIF

cyclotron. It was then transported to the 88-Inch Cyclotron in a lead-lined container, where it was injected into the AECR-U and accelerated. Each batch of ^{11}C was produced in a period of about 50 minutes, separated and transported in 15 minutes, and injected into the source over a period of 15 to 30 minutes. It was during these batch tests that a ^{11}C beam was first accelerated.

Due to the low intensity of radioactive beams, it was necessary to tune the cyclotron optics with stable analogue beams. The cyclotron was initially tuned on $^{22}Ne^{8+}$, then the cyclotron frequency was adjusted to accelerate the trace amounts of residual $^{11}B^{4+}$ always present in the ion source (the cyclotron frequencies for ^{11}B and ^{11}C, with a separation of only 1.4 kHz, are unresolvable). Then ^{11}C was introduced. A scattering foil and particle-identification telescope were used to analyze the resulting accelerated beam. The measured beam intensity was 0.5 to 1 x 10^8 ions/sec, which was maintained over a period of about 20 minutes.

The ^{11}B-to-^{11}C beam ratio was about 1% during the initial test, but increased by more than three orders of magnitude after an unrelated experiment contaminated the AECR-U with boron. However, the boron component of the beam could be eliminated easily by stripping the beam after acceleration and magnetically separating fully-stripped $^{11}C^{6+}$ from $^{11}B^{5+}$.

The ion-source hold-up time for ^{11}C was also investigated. Activity was injected into the AECR-U and extracted as $^{11}C^{4+}$, the current being measured on a Faraday cup after acceleration and stripping to 6+. Figure 1(a) shows the ion-beam intensity-decay curve following interruption of activity injection after less than one minute. There is an initial fast decay time of about 24 s. This component is associated with the holdup time of ions in the plasma and is consistent with measurements for stable CO_2 [6]. A slower component is also observed; it is more clearly seen in Figure 1(b), where the decay curve is shown after 12 min of activity injection. This slow decay, with a mean decay time of about 6 min, is associated with sticking of the plasma-dissociated components of CO_2 to the ion-source plasma-chamber walls.

This batch-mode method was used to perform the first experiment with BEARS: a measurement of cross sections for the production of astatine isotopes by (^{11}C,xn) reactions on a gold target [7].

Transfer Time Tests

The two accelerators in the BEARS system are located more than 300 m apart and a short gas-transport time is required, both to reduce decay losses suffered for short-lived isotopes like 71-sec ^{14}O, and to minimize radiation levels outside the two accelerator buildings. Tests were carried out, without radioactivity, by using bursts of helium gas in an overall flow of nitrogen. The differing response of a thermal-convection vacuum gauge to helium and nitrogen allowed for timing measurements to be made. It was found that the fastest transport times could be achieved by pre-evacuating the transport capillary for a few minutes before applying the pressurized gas. A volume of helium, comparable to the amount needed to fill the BIF gas target, injected in front of the driving nitrogen, traveled the 300-m distance in 12-30 seconds. The exact time depended on the capillary size, the drive-gas pressure, and the degree to which the capillary was pre-evacuated. The regime investigated spanned 2 to 4 mm inner capillary diameter, 1 to 8 ATM pressure, and 1 to 5 minutes pump-out. Capillary evacuation could be improved by supplying drive gas for only a short time, approximately 10 seconds. Longer drive times did not result in noticeably faster transport.

Final BEARS configuration

The following sections describe the entire BEARS system along with its typical operation for the production of ^{11}C beams.

Target System at BIF

Figure 2 shows the BIF gas-target system [1]. The target, 80 mm deep and 13 ml in volume, is filled through valve V1 to 22 atm with the nitrogen target gas. The gas is then

bombarded for 5 minutes with 10 MeV protons, currently with intensities of ~30 μA. During bombardment, the pressure in the water-cooled target increases to 50 atm due to beam heating. After 5 minutes, the beam is shut off and the irradiated gas is unloaded through valve V2 into a "holding tank" where the gas is held prior to transport. After unloading, the target is refilled and the cycle is repeated.

In order to be transported and cryogenically separated by the rest of the BEARS system, the ^{11}C activity must take the chemical form of CO_2. A standard technique used by many PET facilities is to add about 1% oxygen to the nitrogen target gas, typically resulting in better than 60% of the activity being produced in the form of $^{11}CO_2$. However, the use of pure N_2, as was the case for the development tests described above, also results in significant $^{11}CO_2$ yield, possibly due to trace residual oxygen in the gas or chemical reactions on the walls of the target chamber.

Initial BEARS operation used pure nitrogen as the target gas, as the addition of oxygen was found to result in the production, under beam bombardment, of non-radioactive chemical species that were cryogenically trapped along with $^{11}CO_2$. At a 1% oxygen admixture, these gases overloaded the ion-source plasma and seriously degraded the performance of the AECR-U ion source. However, the use of pure nitrogen was found to result in poorer yields of $^{11}CO_2$, less than one half of that possible with the 1% oxygen mixture. In addition, the yields were unreliable, tending to decline slowly during continuous production.

By controlling the exact fraction of oxygen in the nitrogen target gas, a balance was found with both a good $^{11}CO_2$ yield and a gas load low enough for good AECR-U ion source operation. An additional load valve and gas supply was added to the gas-target system shown in Figure 2; the target was first partially loaded with 1% O_2 plus 99% N_2, then topped off to 22 atm with pure nitrogen. The optimum oxygen fraction was found to be around 0.2%.

Transport

Figure 3 illustrates the components of the transport system, which carries the activity from the BIF facility to the 88-Inch cyclotron. While the BIF cyclotron is preparing a 5-min batch of ^{11}C, the transport capillary (polypropylene; 3.0-mm i.d.) is evacuated from both ends, both through valve V7 at the 88-Inch Cyclotron, and through valve V6 at BIF. The holding tank (a 3 m length of 4.5-mm i.d. tubing) is also evacuated at the same time (valves V4 and V6 open, V2 and V3 closed). Valve V4 is closed just before the target is unloaded through V2 and the activated target gas is held in the holding tank for 1 to 2 minutes to allow some of the ^{14}O, not needed during ^{11}C operation, to decay. To transport the gas, 6 atm of nitrogen is applied through valve V3, driving the target gas down the evacuated transport capillary (V2 and V6 closed; V3 and V4 open). The drive gas (V3) is shut off after about 10 seconds, and the pump-out valve (V6) is opened soon after. The approximately three minutes of pumping serve to reduce the pressure in the holding tank to less than 0.2 atm before it is valved off for the next target-unload cycle. Transport between the two buildings takes 20 to 23 seconds. All the activity arrives at the 88-Inch Cyclotron within a spike of about 2 seconds, indicating a lack of mixing with the drive gas.

During most of the 5 minute cycle, the downstream end of the transport capillary is connected directly to the high-capacity pump through three-way valve V7. Simultaneously, the cryogenic trap, a stainless steel coil submerged in liquid nitrogen, is being maintained at vacuum by a second "low-pressure" pump. For about a 10 second period, centered on the expected arrival time of the transported activity, the gas flow is diverted through the trap via valves V7, V8, V9, V10 and then to the high-capacity pump. After the activity arrives, the flow is redirected directly to the high-capacity pump, and the remaining nitrogen gas in the trap is removed by the low-pressure pump through valves V9 and V10.

Figure 4 presents, for a typical transfer, the pressure at the capillary outlet as measured after the trap (this gauge is marked in Fig. 3), and the activity measured in the cryogenic trap by a radiation detector.

Cryogenic System

Before being injected into the AECR-U ion source, the ^{11}C activity must be released from the cryogenic trap by heating. The trap itself consists of a small coil of thin-walled (0.4 mm), 4-mm i.d. stainless-steel tubing. The entire coil is enclosed within a 5-cm vertical tube of low heat-capacity foam insulation. This open-bottomed tube is partially submerged in a liquid-nitrogen bath such that the liquid level completely covers the trap coil. To warm the trap, pressurized room-temperature dry nitrogen is applied to the outer tube, displacing the liquid nitrogen and bubbling out through the bottom of the tube. The flow of gas past the trap coil causes it to warm. A thermocouple, fixed inside a separate section of identical steel tubing, monitors the temperature. With the thin-walled steel tubing currently used, the trap can be warmed from 77 K to 220 K in about 20 seconds.

Ion-source injection

Figure 5 shows a schematic of the BEARS system from the cryogenic trap to the ion source. After most of the nitrogen drive gas has been pumped away through valve V9, warming of the trap is begun. When the temperature of the trap reaches ~120 K, pumping is ceased (V9 closed) and the trap is connected to a "reservoir" section through three-way valve V8. The reservoir is located as close to the ion source as possible, about 3 m from the trap. The reservoir itself is of small volume, formed mostly by the 40 cm^3 internal volume of the thermal convection gauge tube used to monitor the gas pressure.

As the trap continues to warm, ^{11}C activity, believed to be in the form of $^{11}CO_2$, is released from the trap and passes into the reservoir. A slight flow of helium bled into the system at a controlled rate from just in front of valve V9 aids in purging the activity from the trap. When the trap reaches ~230 K, it is disconnected from the reservoir. Pumping through valve V9 is resumed, and the trap is cooled again to liquid nitrogen temperatures in preparation for the next trapping cycle. The transfer is monitored by three PIN-diode radiation detectors: one on

the trap; a second on the reservoir; and a third, attached just after valve V8, that observes the activity flowing in the tubing between the two. Figure 6 illustrates data from a typical transfer, showing the cryotrap temperature and the readings in the three radiation detectors. The activity can be seen leaving the trap, passing through the line, and entering the reservoir.

The gas in the reservoir is bled into the ion source at a controlled rate through a proportional solenoid valve controlled by simple feedback from a thermal convection gauge. This feedback maintains the pressure at the outlet at a set value, as long as sufficient gas is available in the reservoir. Stable operation can be easily maintained for reservoir pressures less than a few torr. The line connecting to the ion source is 4.5-mm i.d. stainless steel and about 3 meters long. The conduction of this line is low enough that, even though the AECR-U ion source operates at pressures of a few 10^{-7} torr, the pressure required at the valve outlet is several millitorr, which is high enough to be measured and controlled by the thermal convection gauge.

Ionization and Acceleration

The controlled injection of activity is crucial to achieve stable operation of the AECR-U ion source. The ^{11}C is ionized, extracted, and accelerated according to the techniques described in the earlier section on development tests. A stripper foil is located in the beam line after the cyclotron and the $^{11}C^{6+}$ beam selected in order to completely eliminate the ^{11}B contamination. Once a small amount of fully stripped carbon has been obtained on an amplified Faraday cup (>2 picoamperes), the AECR-U, injection line, cyclotron, and beam line elements may be fine tuned to maximize the ^{11}C yield on target.

Control System

Operation of the BEARS system is fully automated. Isotope production at the medical cyclotron is controlled by the pre-existing system at BIF. A computer performs the target load, irradiation and unload cycle. A separate control program controls transport, trapping

and injection of the activity into the ion source. The computer running this program accesses the various pressure gauges, radiation detectors, valves and other system parameters via FieldPoint[1], a modular distributed-input/output system. One FieldPoint node is located at the medical cyclotron and a second is located at the 88-Inch Cyclotron. Both nodes are connected in series to the BEARS control computer via a serial communication cable that runs along the transfer line. Although the BEARS and BIF control computers are separate, they synchronize their actions through a pair of handshaking signals.

Radiation Control and Safety Systems

Considerable effort in the BEARS project has been spent on minimizing both routine radiation exposure to personnel and the public, and reducing the risk and severity of any accidental release of activity.

The pressurized gas-handling components at BIF are installed within the cyclotron shielding. Air from inside the shielding is continually sampled and monitored for leaking activity.

The transport and pump-out capillaries in the transfer line are contained inside a 5-cm vacuum hose maintained at pressures less than a Torr. Valve V5, not shown in Fig. 2 but located just after V4 at the entrance to the transfer line, is hardwire interlocked on the pressure in the 5-cm hose, such that, in the event of a leak or break in any part of the 350 meter line, activity transport is immediately halted. The 5-cm hose is itself contained within a strong 15-cm PVC pipe.

At the 88-Inch Cyclotron, all the activity-handling components are at pressures less than an atmosphere during normal operation, and pressure-relief valves, set at a fraction of an atmosphere, are used to prevent accidental pressurization. These relief valves are connected to the main high-capacity pump, and valve V5 is also interlocked to the pressure at the inlet to

[1] FieldPoint is a product of National Instruments (www.ni.com).

this pump. Thus, no leaks from the system are possible without first halting activity transport from BIF.

Transported activity not caught in the cryotrap passes through secondary liquid nitrogen traps and then into the pumps. A small portion of the ^{11}C, probably in the form of ^{11}CO, fails to be trapped. In addition, much of the un-ionized activity that passes through the AECR-U ion source seems to be converted into ^{11}CO. To contain this activity, the exhaust from all the pumps, including that from the vacuum pumps on the source, is temporarily stored in a set of large gas containers while the ^{11}C decays away.

The production and activity-handling systems at BIF and the traps, valves and reservoir sections at the 88-Inch Cyclotron are shielded with concrete and lead. Radiation at some of the pumps can exceed 100 mR/hr at 30 cm, but access to these areas is controlled. Due to the high transport speed of 17 m/s, the time-averaged radiation dose at 30 cm from the unshielded transfer line is less than 1 mR/hr. Parts of the line are buried and shielded; access to the remainder of the line is controlled during operation.

Conclusion

During the first successful test of the full BEARS system an on-target ^{11}C beam of better than 1 x 10^8 ions/sec was achieved at an energy of 120 MeV. This beam was immediately put to use, collecting additional data for the $^{197}Au(^{11}C,xn)$ experiment [7]. Since that time, the BEARS ^{11}C beam has been used in the support of three other experiments: the measurement of ^{11}C fusion/fission on gold and platinum isotopes, ^{11}C+proton elastic scattering, and a comparison of ^{49}Fe yields from ^{40}Ca targets via the $(^{11}C,2n)$ and $(^{11}C,3n)$ reactions. The BEARS system was found to operate smoothly over periods of days with only minor adjustments.

Although currently operating on a 5 minute cycle for ^{11}C, the system is expected to function well at a shorter period, allowing its use with shorter lived isotopes. Work is currently in progress on the development of an ^{14}O beam.

ACKNOWLEDGMENTS

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FIGURE CAPTIONS

FIGURE 1: Beam intensity profile for a $^{11}C^{6+}$ ion beam following activity injection into the ion source for (a) less than one minute and (b) 12 minutes. In both cases the flow of activity into the source was halted at t=0.

FIGURE 2: Target system for activity production at the Biomedical Isotope Facility.

FIGURE 3: System for transporting activity between the two accelerator buildings. Valves are indicated by V2 through V10.

FIGURE 4: Capillary-outlet pressure (solid line) and activity observed in the cryogenic trap (dashed line; arbitrary units) during a typical transfer between the two accelerator buildings. Transport was started (V4 opened) at t=0.

FIGURE 5: System for injection of activity into the AECR-U ion source at a controlled rate.

FIGURE 6: Transfer of ^{11}C activity from the cryogenic trap to the reservoir. The temperature of the trap is plotted in part (a) while part (b) displays the measured activity at three points: the trap (solid line), the reservoir (long-dashed line), and the line connecting the two (short-dashed line).

TABLE 1. Ionization efficiencies of the 88-Inch Cyclotron's two ECR ion sources.

Ion	*ECR*	*AECR-U*	AECR-U with *stable ^{12}C and ^{16}O*
$^{11}C^{1+}$	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 %		
$^{14}O^{5+}$	0.4 %		
$^{14}O^{6+}$		3.6 %	27 %
$^{14}O^{7+}$		1.2 %	6 %
$^{14}O^{8+}$		0.4 %	

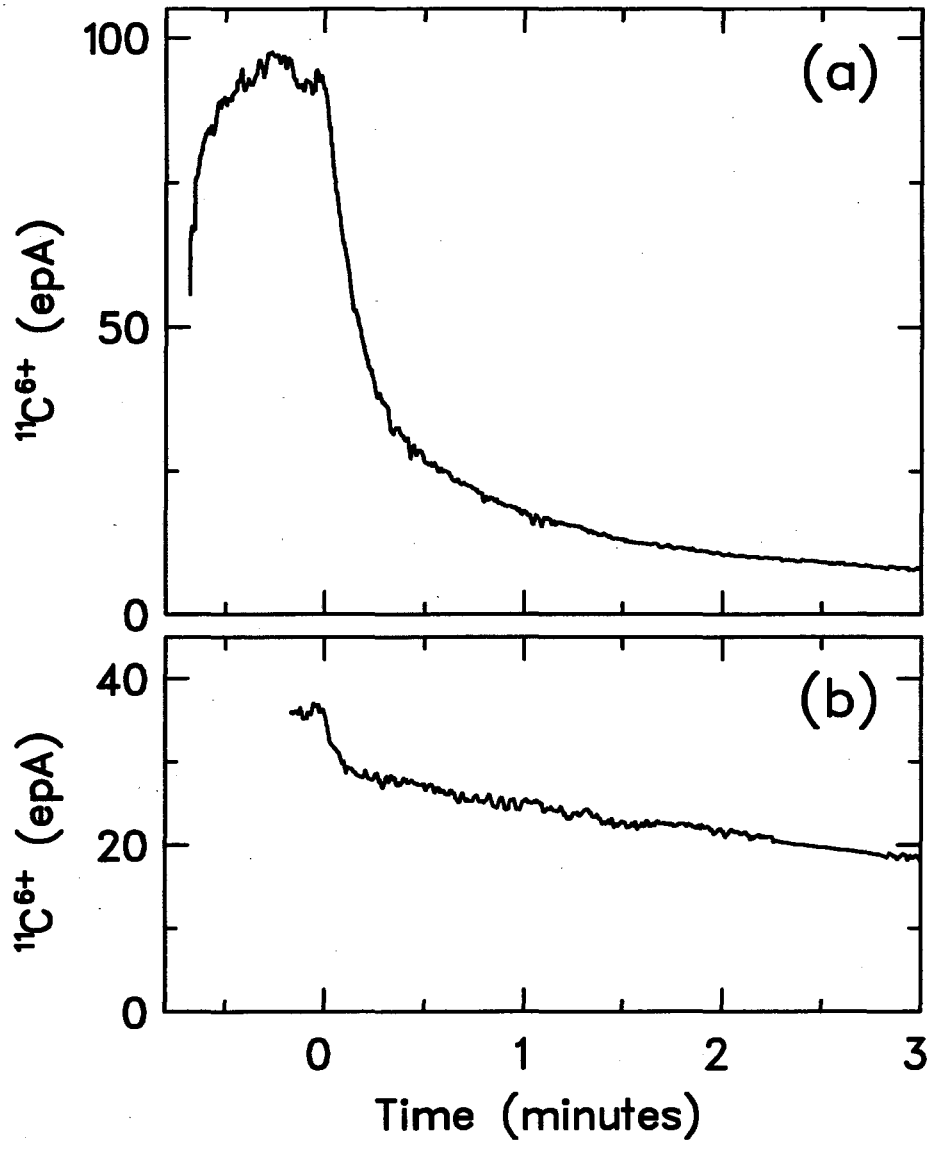
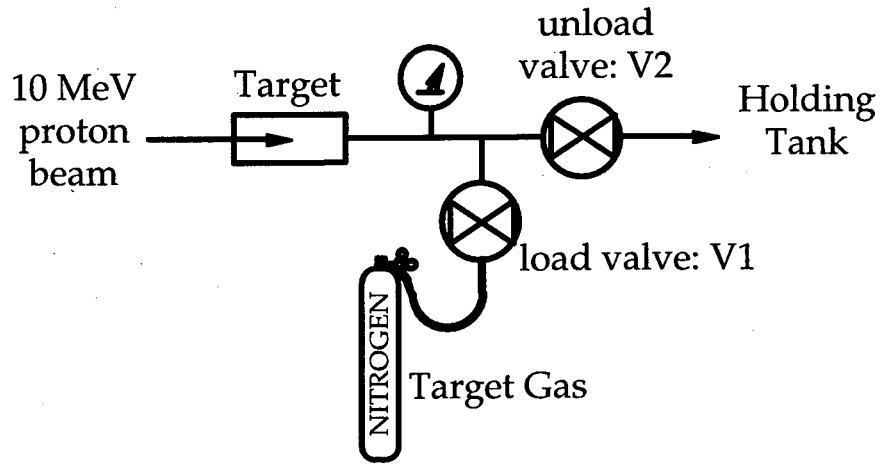


Figure 1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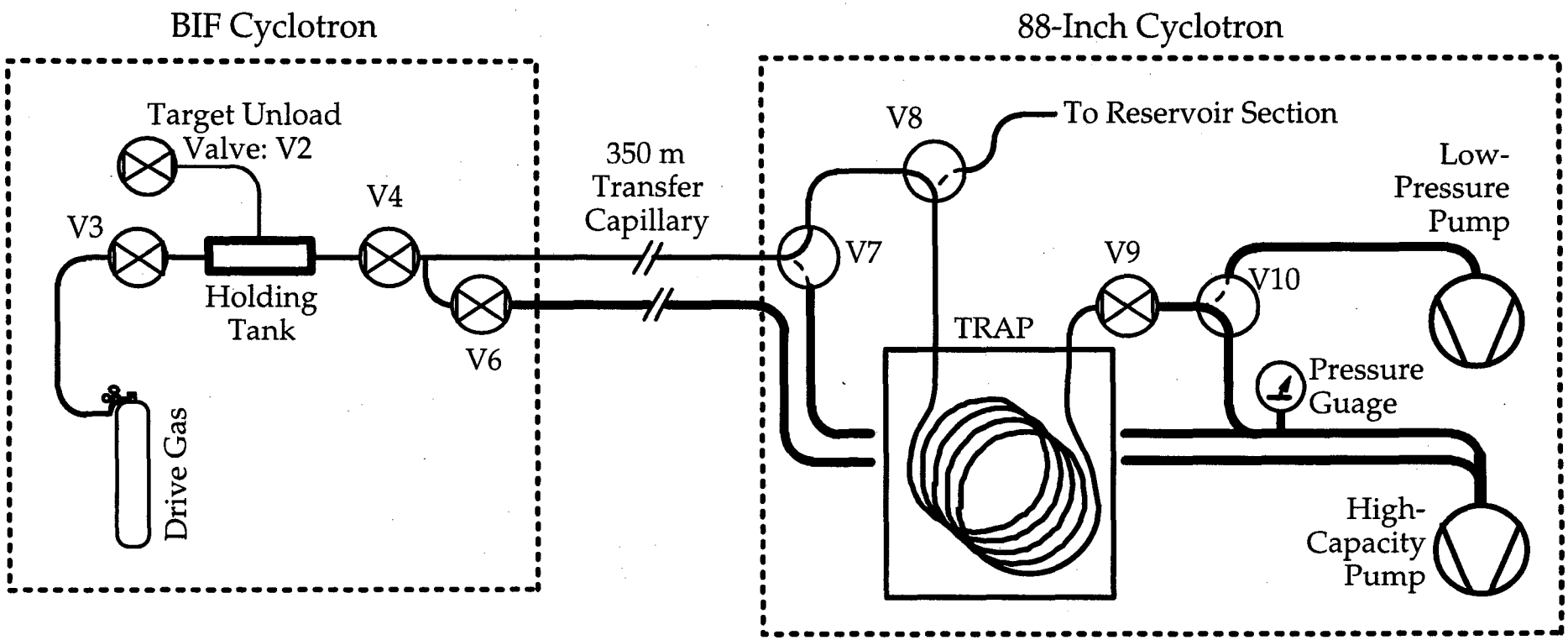
10 MeV
proton
beam
Target
Holding
Tank
load valve: V1
NITROGEN
Target Gas

Figure 2


BIF Cyclotron
88-Inch Cyclotron
Target Unload
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V4
Holding
Tank
V6
Drive Gas
350 m
Transfer
Capillary
V8
To Reservoir Section
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V9
V10
Low-
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TRAP
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Guage
High-
Capacity
Pump

Figure 3

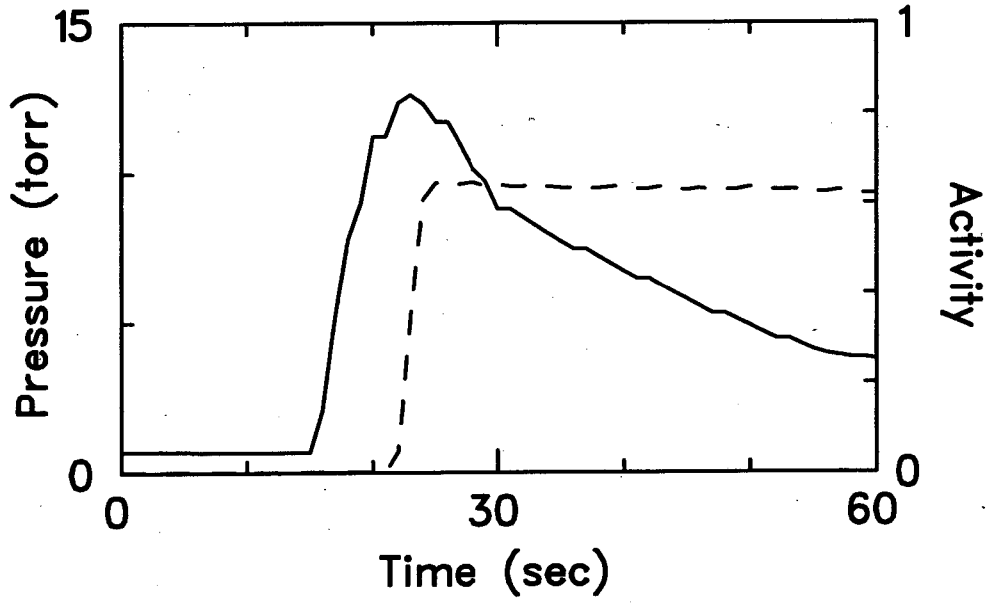

Figure 4

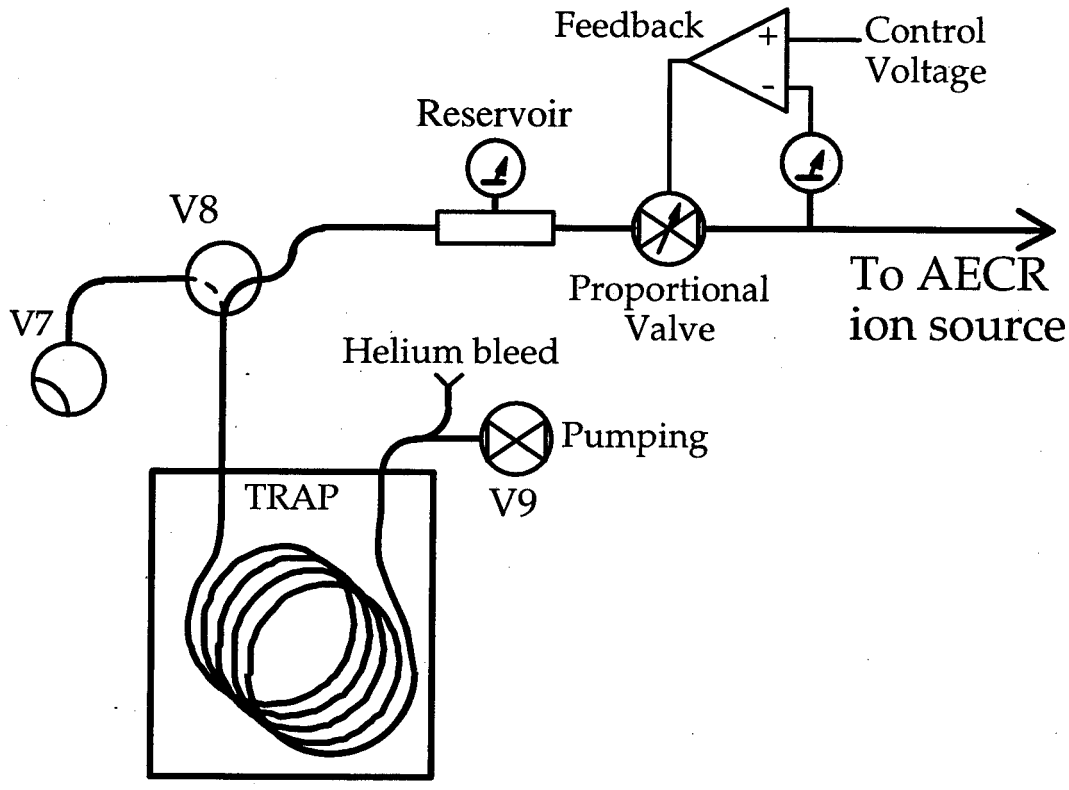
Feedback
+
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Figure 5

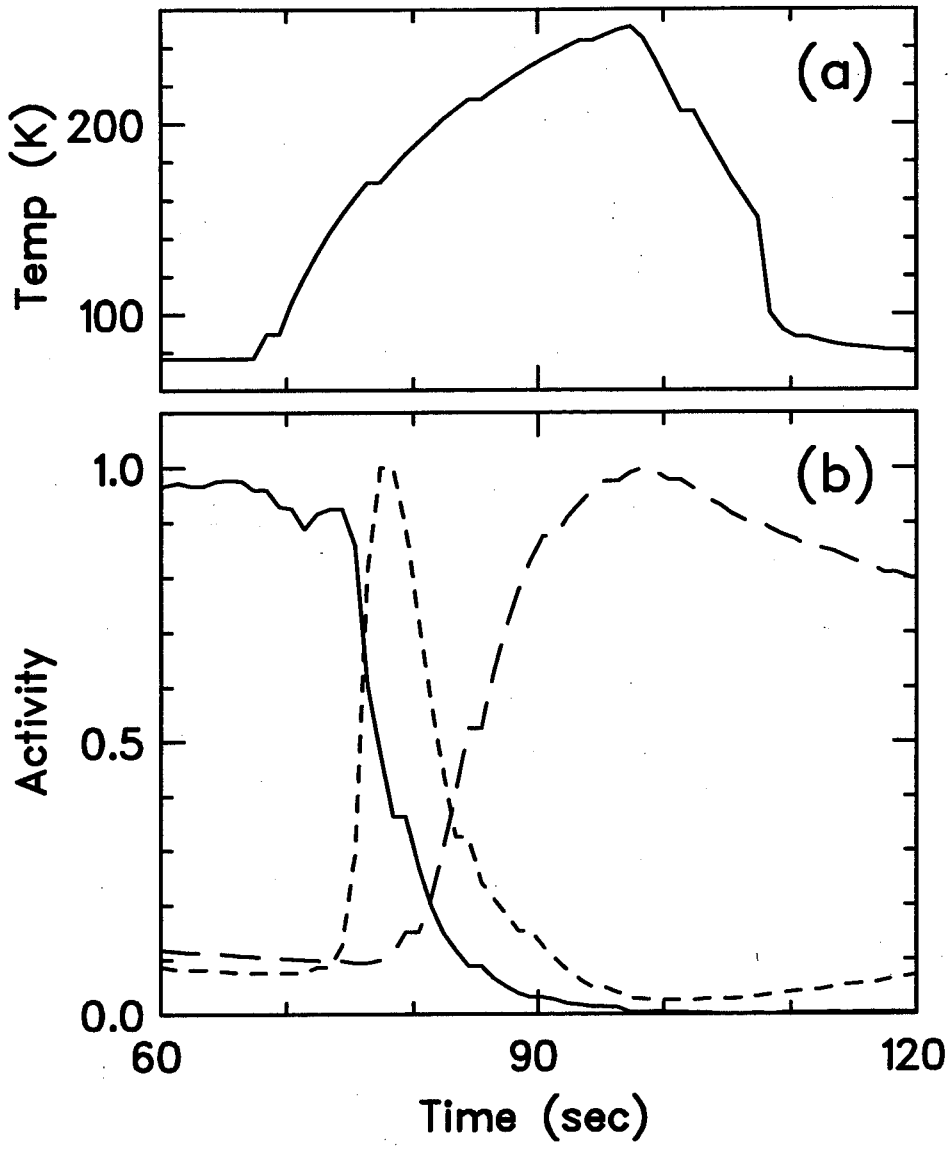

Figure 6

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