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Production of C-14 and stable ion beams at the Argonne Tandem Linac Accelerator System with the ECR3 ion source

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Abstract. The Argonne Tandem Linac Accelerator System (ATLAS) at Argonne National Laboratory (ANL) paused production of carbon-14 ion beams with the removal of the Tandem Van de Graaff in 2013. Installation of ECR3, an Electron Cyclotron Resonance Ion Source, returned that production capability to ATLAS, with the first C-14 beam delivered in December 2020. Information is presented on C-14 beam current, gas consumption and N-14 filtering techniques using stripping foils at different sections of ATLAS. ECR3 also fulfilled the operational goal of adding flexibility to ATLAS stable beam production capabilities. Beneficial impacts to ATLAS operation, beam development and experimental programs are discussed.

1. Introduction

For many years, carbon-14 and stable ion beams were produced at ATLAS using a SNICS II negative ion source coupled with a Tandem Van de Graaff Accelerator to inject positive ions into a linac for further energy gain. The Tandem was removed in 2013 to make room for a stopped beam experimental hall, halting C-14 production. A few years later the former ECRCB which provided stable beams and charge bred radioactive beams from CARIBU was replaced with an EBIS dedicated to charge breeding. With these changes, ATLAS stable beam production was entrusted to a single ion source ECR2. The ECR3 [1,2] ion source was installed to deliver beams of C-14 with the benefit of providing flexibility in stable beam production. High carbon-14 beam purity from an ECR source is intrinsically difficult to achieve because nitrogen-14 has an inseparable mass and is an unavoidable contaminant in ECR ion sources. Techniques developed to filter out N-14 in the ATLAS linac and the resulting target/ion source purity relationship are discussed. C-14 consumption and beam transmission under different accelerator configurations are also examined.

2. Carbon-14 beams from ECR3

ECR3 was commissioned in 2019 using stable beams of He and O. The C-14 would be produced using an ethylene gas with a specific activity of >100mCi/mmol. Experiments were performed with C-13 ethylene to develop baseline production information and to understand C:N ratios under different running conditions [2] before introducing C-14 into the system and complicating operations. First delivery of C-14 to target was achieved in December 2020 with some delay attributed to the COVID-19 outbreak. By conference submission (9/2021), over 30 days of C-14 beam time have been achieved at the 24 hour facility.

2.1. Experimental requirements

The latest approved ATLAS experiments requiring C-14 beams call for beam intensities as high as 300 pnA and energies reaching 210 MeV. These requirements can be satisfied with the flexibility

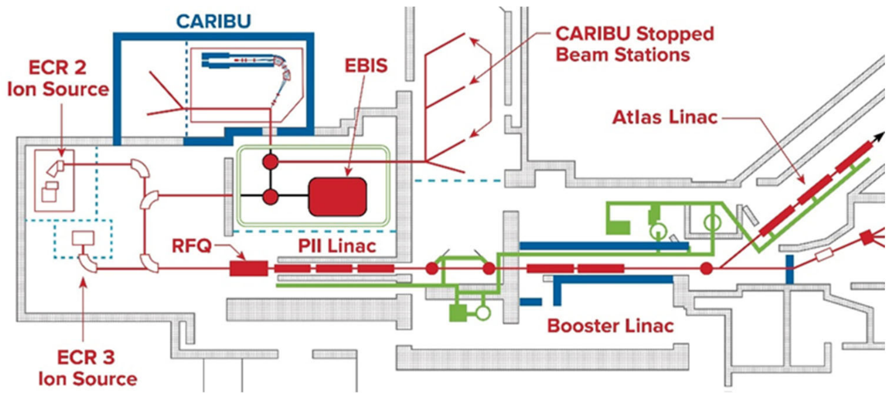

Figure 1. ATLAS front end layout including ion sources and accelerating sections

afforded by the accelerator configuration. Figure 1 shows ATLAS has three distinct accelerating sections: Positive Ion Injector (PII), Booster and Atlas with charge stripping capabilities after PII and Booster. There is a 40 degree bend after Booster allowing m/q selection. The C-13 tests determined that 3+ had the best ionization efficiency and the best LEBT transmission of the charge states that can be accepted by the ATLAS RFQ. Thus for material consumption reasons, unstripped 3+ is accelerated through the machine if the desired energy is achievable. For cases where the 3+ is not sufficient for the required energy, stripping to a higher charge state and achieving more energy gain per linac cavity is required. Table 1 shows the energies achievable with and without stripping under different linac configurations. These numbers tend to be fluid as resonators are brought in and out of service, conditioned for better performance, or de-rated. The highest approved energy of 210 MeV cannot be achieved under the current configuration (A) having 5 resonator cavities awaiting repair and the last cryostat in the Atlas section removed for an energy upgrade scheduled for completion in early 2022. Up to 254 MeV is expected with current configuration combined with the energy upgrade. The energy range would be extended even further with continued resonator repairs approaching 281 MeV (configuration C). A stripping location that yields energy required, high C-14 purity and best possible transmission to target is to be considered for any given experiment.

Table 1. Unstripped and stripped energy of C-14 beams under the following configurations: A) only active resonators turned on, B) active resonators with energy upgrade, C) all resonators with energy upgrade

Entrance charge state			Configuration energy Atlas exit (MeV)		
PII	Booster	Atlas	A	B	C
3	3	3	90	145	165
3	3	6	110	220	242
3	5	5	133	220	246
3	6	6	147	254	281

2.2. Beam stripping

Calculations were performed using LISE^{++} ETACHA4 [3] and in-house semi-empirical based charge distribution codes to approximate stripping fractions using thin carbon foils at PII and Booster energies with results shown in Table 2. Measurements of N-14 stripping fractions were taken using a pure N beam from ECR2. The measured and ETACHA4 fractions for N-14 Booster stripping agreed.

Charge state intensities from an ECR3 mixed beam of C-14 and N-14 were measured after Booster. The N-14 charge state intensities (I_{Nq+}) were determined from the unique N7+ intensity (I_{N7+}) and the measured nitrogen stripping fractions (η_{Nq+}) from the pure beam:

$$I_{Nq+} = I_{N7+}\eta_{Nq+}/\eta_{N7+} \quad (1)$$

Table 2. Calculated and measured stripping fractions of C-14 and N-14. PII exit energy 25 MeV and foil thickness 30 g/cm^2, Booster exit energy 65.7 MeV and foil thickness 75 ugcm^2.

		PII stripping fractions				Booster stripping fractions		
Source	Beam	4+	5+	6+	7+	5+	6+	7+
ETACHA4	C-14	0.03	0.26	0.71	-	0.04	0.96	-
In-house	C-14	0.17	0.47	0.35	-	0.36	0.58	-
ETACHA4	N-14	0.01	0.08	0.37	0.54	0.002	0.068	0.93
In-house	N-14	0.04	0.24	0.45	0.26	0.08	0.38	0.53
Measured	N-14	0.02	0.25	0.53	0.20	0.002	0.088	0.91

The C-14 charge state intensities were found by subtracting the nitrogen component from the mixed beam intensity of a given charge state. The stripping fraction of a given C-14 charge state could then be calculated by dividing its charge intensity by the sum of all C-14 charge intensities. The C6+ fraction was 0.955 and C5+ was .0455 which agree with the ETACHA4 values for carbon.

For stripping the pure N-14 beam at PII energy, the beam transport section was scaled for charge states which were individually identified and tuned in the bend after Booster. The ETACHA4 calculation for PII stripping disagreed considerably with the measured value, possibly due to the lower energy. While results more closely matched the in house code, they were still off by ~18% for N6+ and ~23% for N7+. The PII measurement had limitations. The distance between stripping and analysis is 35m. The stripped charge states were not accelerated in Booster, simplifying the measurement due to time constraints. Scheduling did not allow for a measurement of the mixed ECR3 charge states stripped after PII. A spur is being installed for a new target station 5 meters past the PII exit. It is planned to measure stripping distributions in this location of both pure N-14 from ECR2 and mixed beams from ECR3 upon completion.

2.3. Purity relationship

Stripping energy and the charge state being selected have an impact on beam purity at target. We define the beam purity as the percentage of carbon-14 in the mixed beam with the remaining being nitrogen-14, inseparable by m/q. A beam purity of 80% or greater (C:N ≥ 4:1) is the most stringent requirement of the currently approved experiments. The effect stripping has on beam purity by filtering out nitrogen can be derived mathematically with stripping fractions at a given energy:

$$P_{tgt} = \frac{P_{src}\eta_C}{P_{src}\eta_C + \eta_N(1\text{-}P_{src})} \tag{2}$$

where P_{tgt} and P_{src} are the beam purities at target and ion source. η_C and η_N are stripping fractions of the same charge state for carbon and nitrogen respectively. Figure 2 plots this equation for mixed beams stripped at PII and Booster with energies and foil thicknesses used in Table 2. The PII error

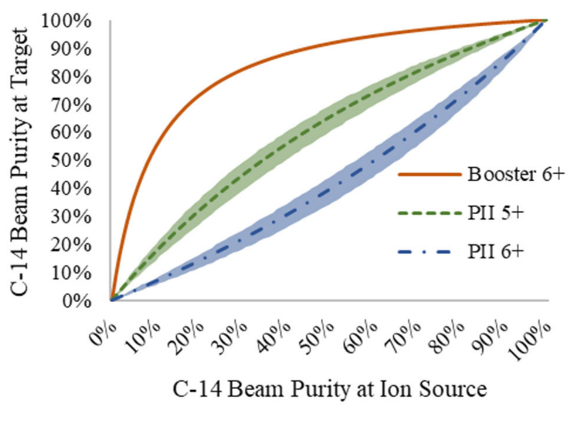

Figure 2. Target and ion source purity relationship for beams stripped to different charge states at Booster and PII.

bands come from +/- 20% C-14 stripping fraction uncertainty from using the in house code. Obtaining >80% target purity is most easily achieved by not stripping at PII and stripping to 6+ at booster, followed by stripping to 5+ at PII and not stripping at Booster. Stripping to 6+ at PII results in target purity that is worse than at the ion source.

2.4. C-14 experiment runs

Table 3 displays a series of nuclear physics experiments in order of operation that were run at ATLAS using different stripping schemes over the past year. Consumption rate was determined with pressure readings taken before and after a run of the fixed volume of gas using an Omega DPG2001B [4] pressure gauge. The C-14 transmission was determined by multiplying the C-14 purity and mixed beam intensity at the last common faraday cup before target stations and dividing by the same product at the source cup. Attenuation was also accounted for. Transmission varied over the course of a run due to progression of tuning, foil lifetime and changes in source purity.

Table 3. Carbon-14 experiments at ATLAS

		Beam parameters at target			Charge state		C-14	
Experiment number	Hours	Intensity (pnA)	Energy (MeV)	Purity requested	Booster	Atlas	Cons. rate (mg/hr)	Transmission
1728	88	1	140	~50%	6+	6+	0.025	8.5-12%
1739a	137	100	122	> 80%	3+	6+	0.075	36-45%
1732	165	16	100	> 80%	3+	6+	0	23-31%
1739b	216	95	122	> 80%	3+	6+	0	36-40%
1955	160	100	133	> 80%	5+	5+	0.16	6-14%

The first experiment, 1728 required a mixed C-14/N-14 beam. Pure nitrogen beam from background gas was delivered to target for characterization, then C-14 was introduced until beam current at the source doubled. A purity of ~40% was verified from sampled ionization rate and total beam rate in an ionization chamber [5] and from elastic scattering (Rutherford regime) in a recoil E-dE silicon detector at the end station. Stripping was done at PII exit to reach energy and a factor of 5 attenuation was used to reach requested intensity.

The following experiment, 1739a was able to reach energy by stripping at Booster exit with a C-14 consumption rate of .075 mg/hr while delivering 100pnA. With a higher stripping fraction into 6+, transmission was improved from the previous run. For experiment 1732, the source was run at 7 watts while delivering 16 pnA on average to target without introducing C-14 gas. The drop in transmission is attributed to tuning required to reach a farther target station. Experiment 1739b immediately followed. We were able to increase output with an RF increase to 14W. Helium support was increased occasionally over the run to maintain ~95 pnA on target, also without C-14 gas feed. Remarkably, both experiments combined for 381 hours without gas feed. This can be attributed to wall recycling of the ethylene gas introduced previously. During the runs the target purity steadily dropped from 91% to 85% while the source purity fell from 48% to 34% indicating the usage of material from the walls. The effective consumption rate over 518 hours was 0.02 mg/hr.

Unfortunately the energy goal was not met with Booster stripping for experiment 1955. Poor C-14 transmission due to the PII stripping fraction into 5+ resulted in a return to active gas feed with higher consumption than previous to maintain intensity. It is possible that material from this run may be recycled during future runs depending on stable beam demands from this source.

3. ATLAS operational relief

As previously noted [1,2] the ECR3 installation has another benefit that cannot be overlooked. Aside from providing the C-14 beam which was lost to ATLAS in 2013, ECR3 has demonstrated that it can readily provide stable beams of z = 1 to 36 originating from gas. Between commissioning in October 2019 and September 2021, 71 operational and tune days were allotted to ECR3. Many of these days were used concurrently for ion source development and maintenance at ECR2. Without ECR3, these activities would have caused reduced programmatic hours or been deferred. Figure 3 shows a historical allotment of ion sources used for the production of stable beams. In the years leading up to 2019, ECR2 became the solitary source of stable beams at ATLAS. ECR3 produced 8% of the stable beam hours delivered this past year. Another 29% of those hours were readily producible from ECR3 but ran from ECR2 instead. With more operational knowledge and experience gained ECR3 can be used for more stable beams in the future.

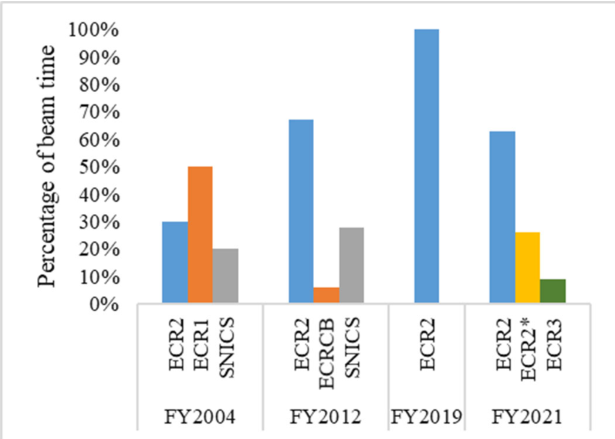

Figure 3. Sampling of ATLAS ion source usage for stable beams over time. *ECR2 beams that ECR3 is capable of producing.

4. Summary

The ECR3 ion source has provided C-14 beams while adding flexibility to the ATLAS experimental program as a backup and complementary source to ECR2 for stable beam production. Nitrogen filtering using a stripping foil has been demonstrated. From material consumption and beam purity standpoints, the ideal configuration has been found to be stripping to 6+ at Booster when energy cannot be achieved unstripped. After the pending energy upgrade of ATLAS, most experiments will use this configuration. Wall recycling of the ethylene material was also demonstrated without active gas feed, which can reduce the overall consumption rate. A wish to conserve material may conflict with the desire to use the source for stable beams since a part of the remaining C-14 would be consumed during operation. Strategic timing of experiments may provide a novel solution.

Acknowledgments

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