30TH CONFERENCE OF THE
INTERNATIONAL NUCLEAR TARGET DEVE

THE PREPARATION OF ISOTOPIC BORON TARGETS – SEARCHING FOR A MORE CONSISTENT APPROACH

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ABSTRACT

The reliable availability of isotopic boron targets has gained in importance concerning light ion reactions withing the realm of nuclear astrophysics research and still remain somewhat elusive. An exhaustive approach was undertaken via electron beam evaporation to produce self-supporting and backed targets spanning various isotopic samples across a number of suppliers, resulting only in limited success. Details of the extensive sample preparation procedures undertaken, and the experimental deposition techniques explored will be presented.

Legacy Boron samples, natural and isotope

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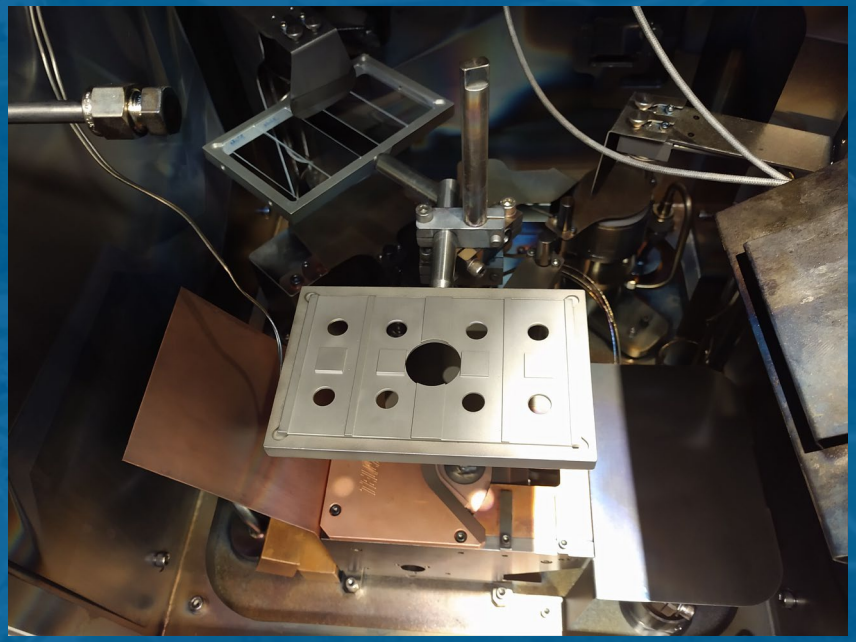
OUTLINE OF THE TALK

- Introduction & Motivation
- Review of Previous Work
- Isotope and Sample Preparation
- Experimental Procedures
- Results & Discussion
- Outlook & Conclusion



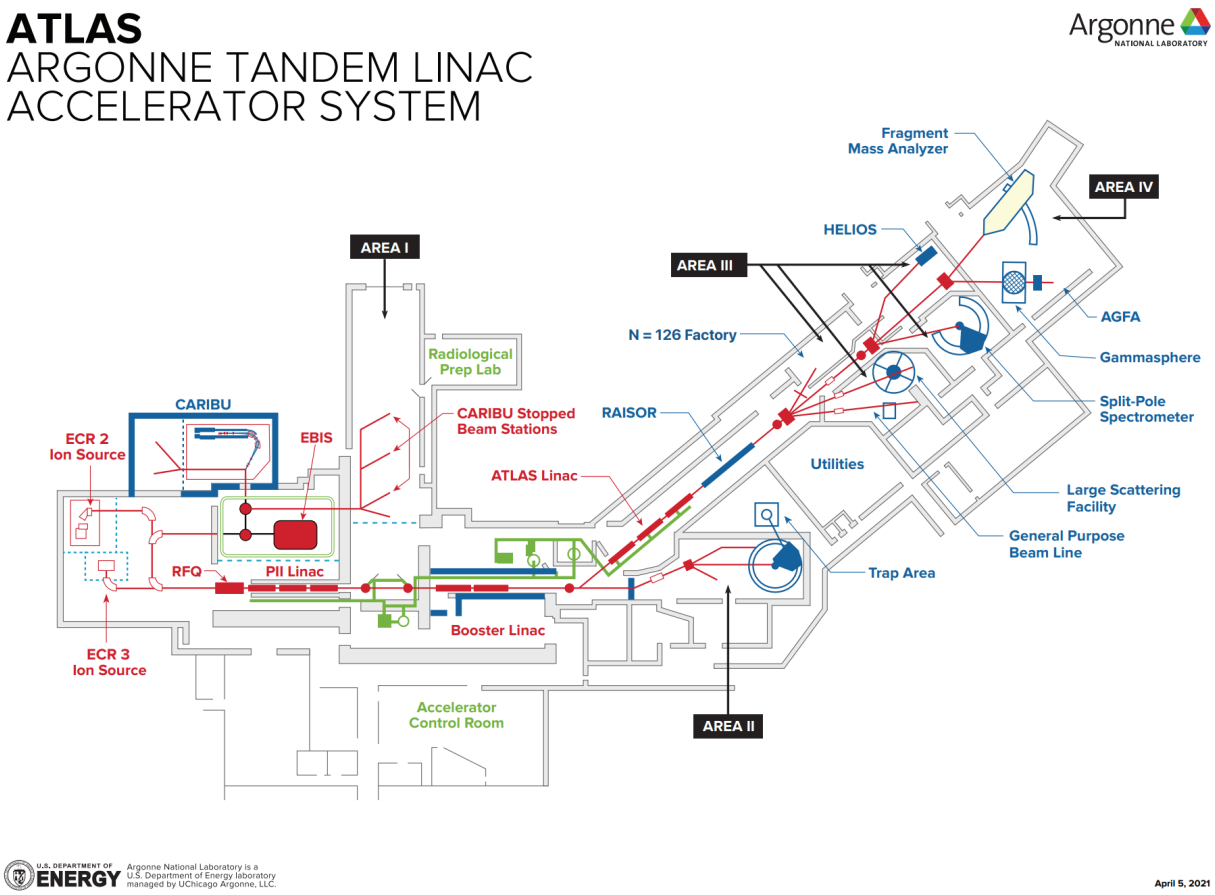
Deposition set-up for the NaCl substrates (rear) and carbon backing foils (front)

INTRODUCTION AND MOTIVATION

- For ongoing experiments in low-energy nuclear physics at the Argonne National Laboratory (ANL) ATLAS Accelerator Facility and especially studies involving nuclear astrophysics, isotopic boron targets are becoming more in demand. While current preparation techniques involve Physical Vapor Deposition (PVD) employing electron beam heating [515], reliable and consistent production of targets has been less than successful. Our approach here has been to discover and document a standard procedure capable of consistently obtaining the targets needed for meeting the experimental demands.
- Therefore, a reproducible and reliable preparation of thin self-supporting isotopic boron targets for low-energy nuclear physics applications needs to be researched and developed. We present here a systematic study of an experimental technique using PVD, employing electron beam heating, for isotopic boron thin films to be used as accelerator targets.

INTRODUCTION AND MOTIVATION

ATLAS Facility Layout with Beam Locations



REVIEW OF PREVIOUS WORK

Literature Search – Boron Target Preparation

- There are many references on research pertaining to preparing boron films via the process of electron beam heating, beginning with Eschbach in 1972 as well as others. The results presented here extend the earlier work at ANL of Thomas. Various alternate methods also appear in the literature, including electrodeposition by Verdingh and Pauwels, focused ion beam sputtering by Baumann and Wirth, and also Maier and Muggleton. Xu and Wang employed the method of centrifugal precipitation and Lozowski and Hudson prepared thick targets via powder pressing. A new technique HIVIPP, developed by Sugai was used to produce boron films and also later by Lipski, et al. It should be noted that early on, boron was being considered as an alternative to carbon for use as stripper foils by many researchers, including Ramsay, and with Zeisler and Jaggi, incorporating multi-layers and via laser ablation deposition.
- Finally, a lot of effort has gone into the characterization and assay of boron film layers. For instance, as neutron dosimeters, at IRMM as reference standards for nuclear measurements, and for neutron physics experiments at NIST.

ISOTOPE AND SAMPLE PREPARATION

Boron, B (at. no. 5) (at. wt. 10.81)

- Boron is found in nature as compounds, primarily borax, the element being discovered in 1808 by Sir Humphry Davy. High-purity crystalline boron is obtained from the vapor phase reduction of boron trichloride with hydrogen on electrically heated filaments or via exchange distillation reactions using a boron trifluoride compound *(U.S. Patent 3,050,367).* Its melting point is 2300 C but it sublimes at 2550 C. Boron exists as two isotopes, ^{10}B with 19.78% abundance and ^{11}B with 80.22 % abundance. The isotopes are available as crystalline form in high-purity from ORNL (*www.ornl.gov/facility/redc*) – originally Eagle-Picher (*www.eaglepicher.com*) EP Boron Quapaw, Oklahoma.

Crystalline natural Boron

ISOTOPE AND SAMPLE PREPARATION

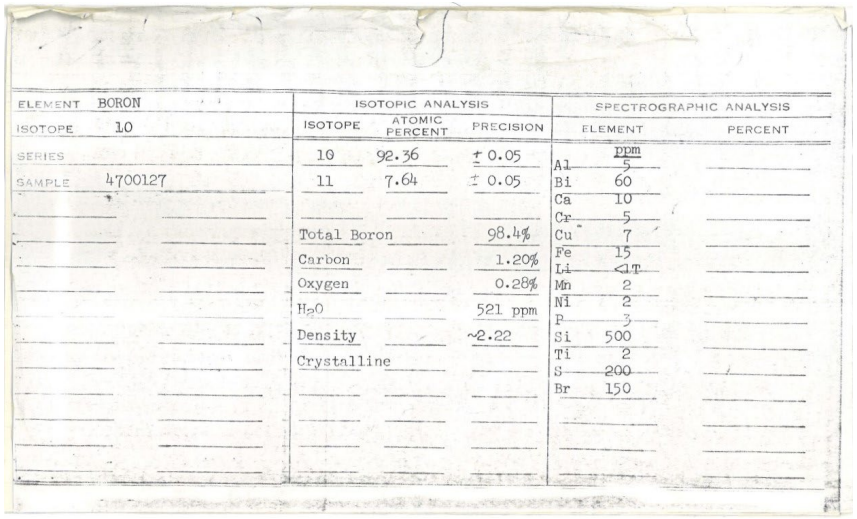
Sample Preparation

- There exists at our disposal several legacy batch samples of various enrichments for both isotopes for use in preparing accelerator targets for ATLAS. Also, a sample of ^{nat}B powder (purity 99.995%) from Lieco Industries (*www.liecoind.com*) available to be used for the initial test depositions.

Legacy Boron samples, natural and numerous isotopic, from various suppliers

ISOTOPE AND SAMPLE PREPARATION

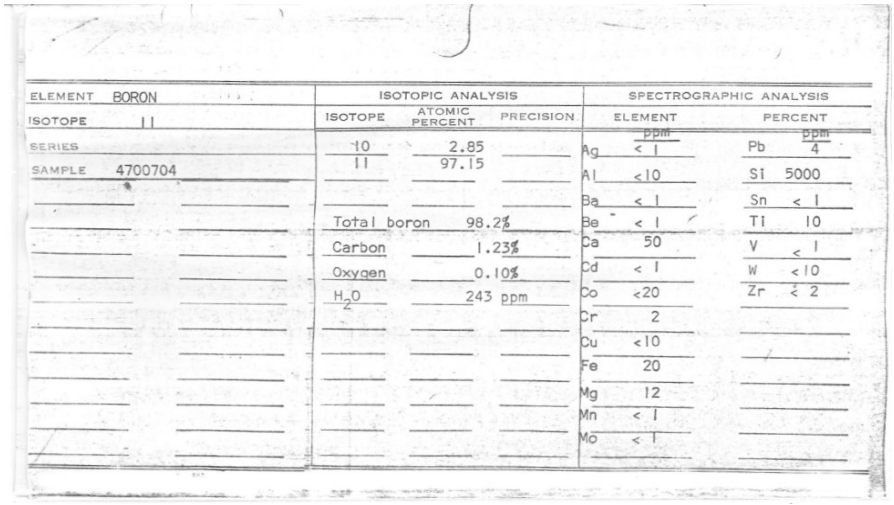
Typical Isotopic Analysis (ORNL ASSAY)



ELEMENT BORON
ISOTOPE 10
SERIES
SAMPLE 4700127

ISOTOPIC ANALYSIS		
ISOTOPE	ATOMIC PERCENT	PRECISION
10	92.36	± 0.05
11	7.64	± 0.05
Total Boron		98.4%
Carbon		1.20%
Oxygen		0.28%
H_2O		521 ppm
Density		~2.22
Crystalline		

SPECTROGRAPHIC ANALYSIS	
ELEMENT	PERCENT
	ppm
Al	5
Bi	60
Ca	10
Cr	5
Cu	7
Fe	15
Li	<1T
Mn	2
Ni	2
P	3
Si	500
Ti	2
S	200
Br	150



ELEMENT BORON
ISOTOPE 11
SERIES
SAMPLE 4700704

ISOTOPIC ANALYSIS		
ISOTOPE	ATOMIC PERCENT	PRECISION
10	2.85	
11	97.15	
Total boron		98.2%
Carbon		1.23%
Oxygen		0.10%
H_2O		243 ppm

SPECTROGRAPHIC ANALYSIS			
ELEMENT	PERCENT		
	ppm		ppm
Ag	< 1	Pb	4
Al	<10	Si	5000
Ba	< 1	Sn	< 1
Be	< 1	Ti	10
Ca	50	V	< 1
Cd	< 1	W	<10
Co	<20	Zr	< 2
Cr	2		
Cu	<10		
Fe	20		
Mg	12		
Mn	< 1		
Mo	< 1		

Note the > 1% carbon impurity (boron carbide?) and also significent silicon

ISOTOPE AND SAMPLE PREPARATION

Behavior of the Isotope – Impurities Present

- As the natural material behaves in a fashion completely different from the isotopes, a further discussion is provided. Most experienced target practioners are quite aware of this behavior. This is particularly true for the case of crystalline boron isotope due to the chemistry involved in the enrichment process. It is exhibited in the fact that boron when heated tends to eject from the electron beam hearth – due presumably to some unknown volatile compound decomposition which is contained in the solid isotope. The question now becomes how to identify and alleviate the problem.

ISOTOPE AND SAMPLE PREPARATION

Pre-Heating to Remove Impurities

- As per the earlier work of Thomas at ANL, the powdered starting material is first pre-heated in an effort to drive out any impurities. This heating was carried out withing the Intlvac Evaporator System using a Ta crucible heated resistively from a crucible oven source. The powder was first pressed into a 0.95 cm diameter pellet using a hand press. The source is brought to approximately 1296 C, measured using an optical pyrometer and heated until outgassing ceases (as registered by the nominal system pressure), and stopped when deposition commences. The pellet was left to cool. It had been weighed prior to the heating and then re-weighed subsequent to removal afterwards. The results of several such heated samples was that most of these powdered samples lost on the order of 15% by weight.

Intlevac Nanochrome deposition system

*NIMA **561** (2006) 58-61*

Parr Pellet Press for compressing powdered samples into pellet form

ISOTOPE AND SAMPLE PREPARATION

Pre-Heating under Vacuum (INTLVAC Deposition System)

SAMPLE	Pellet Diam. (cm)	Weight Before (mg)	Weight After (mg)	% Weight Loss
^{nat}B	0.95	109	90	17
^{nat}B	0.95	202	180	11
	0.64	88	81	8
^{nat}B	0.95	203	177	13
		202	176	13
		199	172	14
^{10}B SS 1(a) 140 1210-1	---	310	291	6
^{10}B #4700117 24-3177	0.95	428	385	10

ISOTOPE AND SAMPLE PREPARATION

Sample Preparation – Ball Milling

- Another aspect of standardization of the boron starting material is particle size. Characterizing the particle size analytically can be accomplished using the electron microscope at CATS, however it's an involved process. Using instead a calibrated set of sieves we can to a first approximation prepare samples with a particle size of 60 mesh (upper limit). This technique works fine for powdered samples, the ^{nat}B powder used was found to be finer (80 mesh). However, the majority of the isotopic material is of crystalline form. To prepare uniform powder starting samples from this crystalline isotope, the large crystalline chunks are first crushed using a percussion mortar and pestle. Next, this material is further reduced in particle size using a SPEX Sample Prep ball mill and Ta ball media. For ALL samples, the resulting powder sample is then passed through a 60 mesh sieve, pressed into a pellet and pre-heated to remove impurities.

SPEX Sample Prep ball mill and media (below)

ISOTOPE AND SAMPLE PREPARATION

Pre-Heating the Powdered Samples in the Tube Furnace under Argon

- The results of the ^{nat}B crystalline sample from Eagle-Picher showed little to no weight loss after vacuum heating. Therefore, as an alternate technique, and in order to avoid evaporation during this heating cycle, the isotopic sample powder pre-heating was carried out in the target laboratory tube furnace under flowing Ar gas. As before, the isotope powder was compacted into a pellet and heated to 1000 C for 1 hour in a Ta crucible. The samples were weighed before and after.

Above - laboratory tube furnace used for preheating boron isotopic samples.

Below – close-up of sample in Ta crucible.

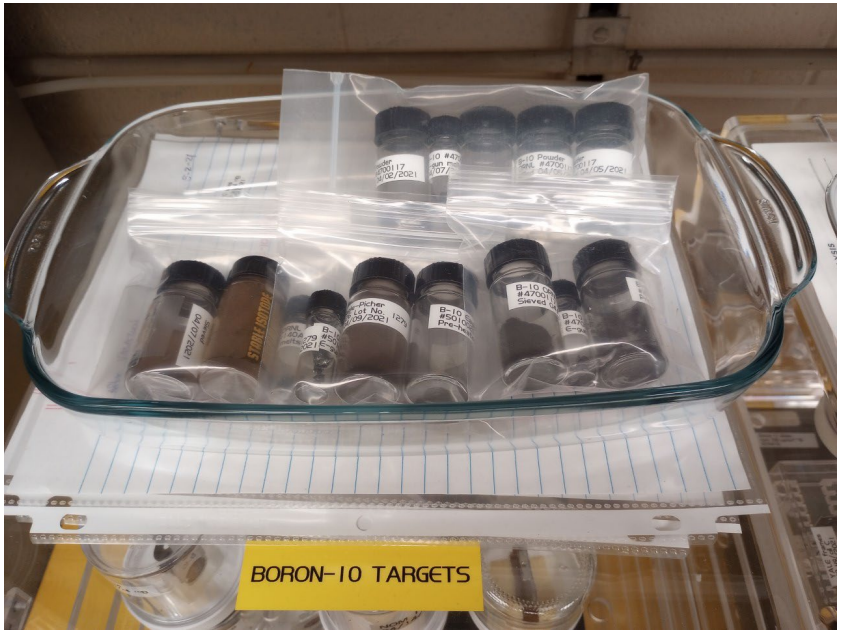
ISOTOPE AND SAMPLE PREPARATION

Sample Pre-Heating in Tube Furnace (1000 C for 1 Hour)

ISOTOPE	Sample Designation	Weight Before (mg)	Weight After (mg)	% Weight Loss
^{10}B	Eagle-Picher 1279 5010026	406	380	6
^{10}B	#4700117 24-3177	407	332	18
		403	324	20
^{10}B	SS 1a-140c 29-1977	408	395	3
^{11}B	SS 2(ce) Req. 1506-204	398	370	7
		403	371	8
^{11}B	ENGLAND #22 20th Century Electronics	407	377	7
		407	382	6
^{11}B	Eagle-Picher B-11-85-1	409	405	1
		397	394	<1

ISOTOPE AND SAMPLE PREPARATION

Isotopic Samples to be used for Target Deposition

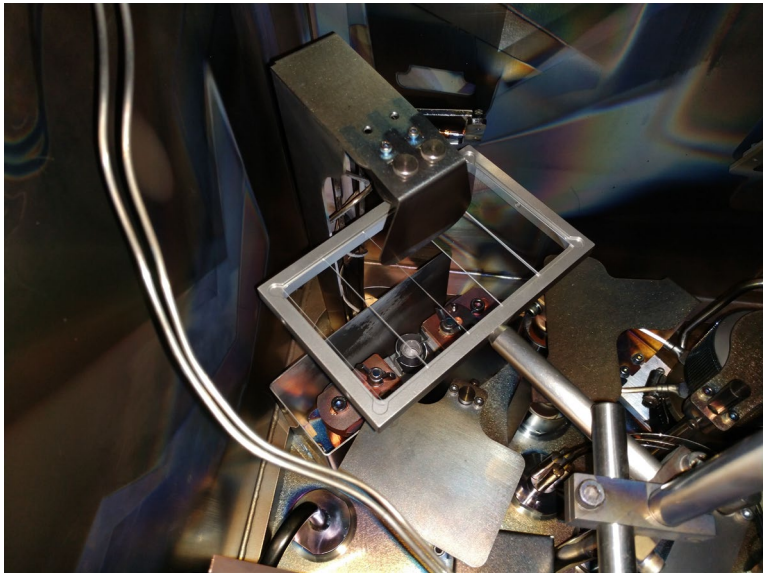


Collections of various milled, sieved and ,melted isotopic boron samples

EXPERIMENTAL PROCEDURES

Substrate Preparation

- The substrates chosen for this work are standard laboratory microscope slides, unfrosted, with dimensions 2.5 cm x 7.5 cm, obtained from Fisher Scientific Inc. Approximately 50-60 μg/cm^2 of NaCl are evaporated onto the slides from a Ta crucible in a resistive heating crucible oven source, also contained within the Angstrom Deposition Tool. From past experience, sodium chloride was chosen over the boron oxide as suggested by Ramsay. For this substrate layer, four clean glass slides are placed above the evaporation source at a distance of 11 cm. The quartz crystal deposition monitor was positioned at 17 cm away, centered directly above the Ta crucible source.



Set-up for substrate evaporation of NaCl

Interior of Angstrom Deposition Tool

(https://doi.org/10.1063/1.5035535)

EXPERIMENTAL PROCEDURES

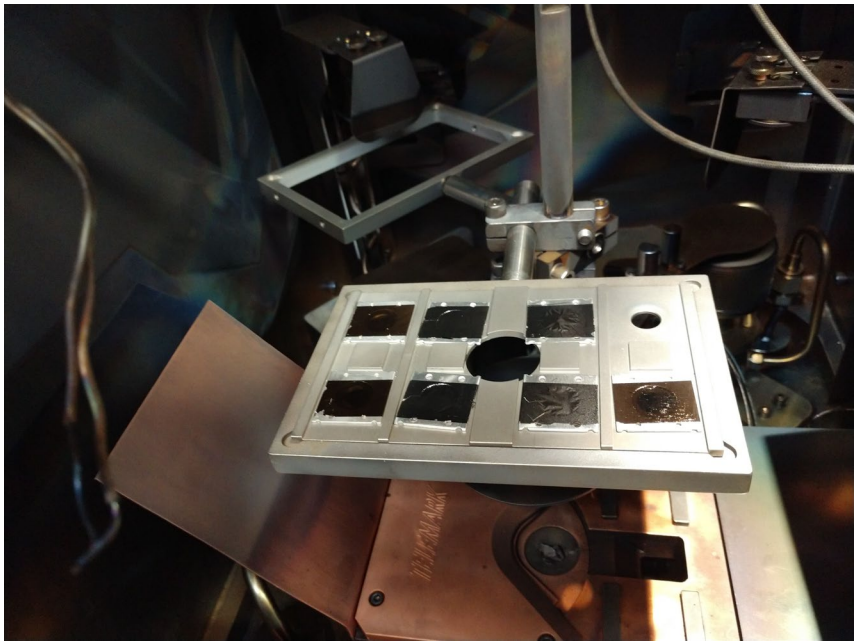
Boron Evaporation from the Electron Beam Source

- The boron evaporations (natural and isotopic) were carried out via Physical Vapor Deposition (PVD) within the Angstrom Deposition Tool using the Telemark electron beam source. Four previously prepared NaCl coated glass substrates were placed at 12 cm distance from the source and are directly heated from a halogen quartz lamp placed above. The substrates are held at a temperature of 176 C, maintained and monitored via a thermocouple placed on the back of the substrate slides. The boron sample is placed in one of the four available Cu hearths of the electron beam source, having first been recompacted into a fresh pellet via hydraulically pressing under vacuum to reduce outgassing.
- For these depositions an arbitrary end-point of 100 μg/cm^2 target thickness was chosen to begin as these would provide usable targets. Thicker depositions could be performed later once the standardized parameters for this work had become established. Once under vacuum and the substrate heating achieved the electron beam power is slowly increased. With a beam voltage of 8-9 kV a deposition rate of 0.2 kÅ/sec was slowly achieved and maintained throughout as measured by the quartz crystal monitor placed to the side at a fixed 22 cm distance from the source. Deposition was stopped when the end-point was reached and the system allowed to cool, usually overnight, before the slides were extracted.

EXPERIMENTAL PROCEDURES

Boron Target Evaporation onto Carbon Backing Foils

- Due to the limited success with producing self-supporting targets, further deposition attempts were then continued on carbon backing foils. Carbon backings of 20,30 and 40 $\mu g/cm^2$ were prepared on the various target frames anticipated as being needed. They were made using the standard floating technique from carbon slides obtained from Arizona Carbon Foil Company as they have proven over time to be a consistently reliable source of high-quality carbon foils for use in our target laboratory.
- The deposition parameters were kept similar to those used for the self-supporting boron films.



Angstrom Deposition Tool set-up for evaporating boron onto carbon backings

RESULTS AND DISCUSSION

Boron Targets Produced

- The resulting efforts from this work include the preparation of many targets for use in experiments at ATLAS and elsewhere. Certainly, the work beginning with ^{nat}B showed that the techniques and sample preparation yielded instantly many targets without problems. It was primarily due to the difficulties arising from working with the isotopic materials that the problems encountered with being able to consistently preparing isotopic targets that prompted this renewed effort to standardize the technique.
- Issues still remain however and further investigations into understanding possible impurities in the 10,11B isotopic material need to be followed up. This work is continuing.

RESULTS AND DISCUSSION

Isotopic Boron Self-Supporting Targets

ISOTOPE	Sample Designation	Thickness ($\mu g/cm^2$)	No. of Targets
^{10}B	SS 1(a) 140 1210-1	110	1
^{10}B	#4700117 No. 24-3177	56	4
^{10}B	Eagle-Picher #5010026 Lot No. 1279	101	12
^{10}B	SS 1a-140c No. 29-1977	83	3
^{11}B	England 20th Century Electronics	101	2
^{11}B	Eagle-Picher B-11-85-1	61	2

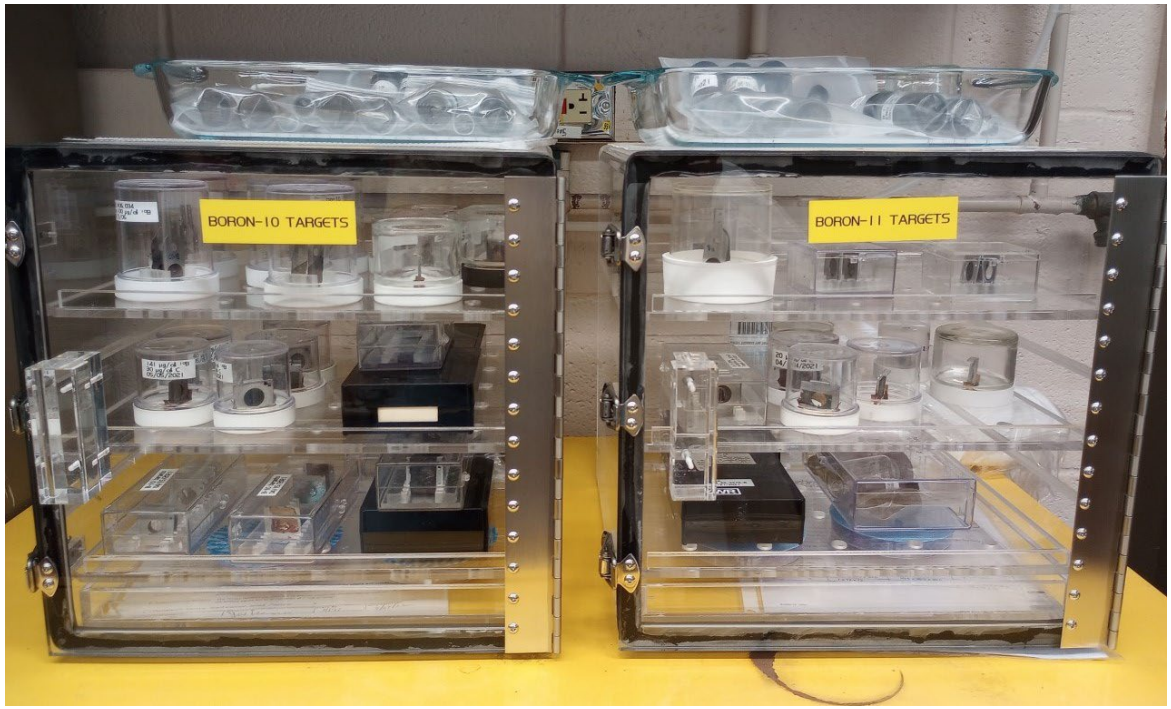
RESULTS AND DISCUSSION

Isotopic Boron Targets on Carbon Backings

ISOTOPE	Sample Designation	Thickness (μg/cm^2)	C Backing (μg/cm^2)	No. of Targets
^{10}B	#4700117 24-3177	141	30	2
		137	40	1
		113	40	1
		107	40	1
		84	30	2
^{11}B	#4700704 No. 26-2578-B	121	40	8
^{11}B	Eagle-Picher Lot B-11-85-1	81	40	5

OUTLOOK FOR CONTINUING WORK

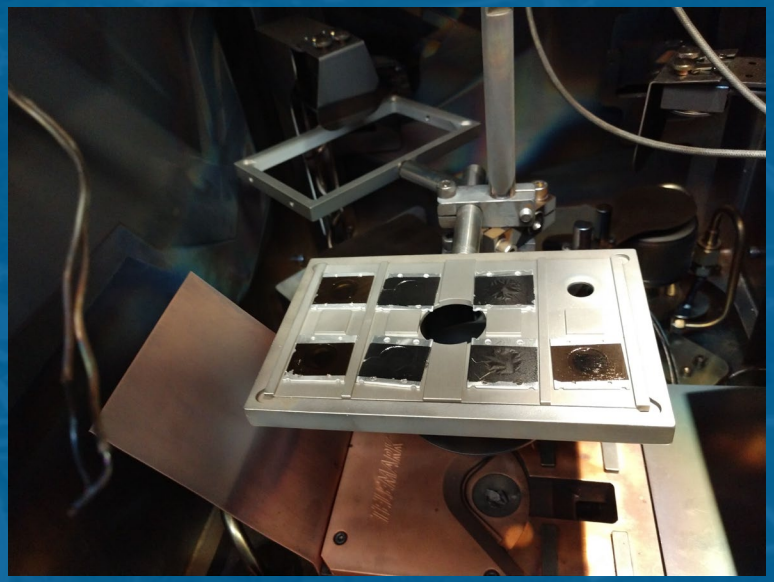
- The limited success outlined here means there is still progress to be sought out. Particle size and powder grinding has provided a good approach, but the difficulties uncovered during target preparation imply the presence of impurities remain. The possibility of sintering the powder at high temperature in a vacuum furnace may provide an avenue for this impurity removal.



Resulting collections of isotopic boron targets and slides

CONCLUSION

In conclusion, although many isotopic sample preparation techniques were explored for both isotopes, no direct correlation was ascertained as to successful target preparation. It still appears that possible chemical compound impurities exist at a level whereby self-supporting films remain difficult to impossible to achieve. It was desired that among the many isotopic samples varying degrees of impurities could be minimized and procedures developed to reduce their behavior on thin film production properties. We anticipate a steady demand for these targets once reliable preparation techniques can be established. Further investigations are pending.



Angstrom Deposition Tool set-up for evaporating boron onto carbon backings

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