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Inferred Effects of Water on Electron Drift Lifetime in Liquid Argon

Abstract

A material test system (MTS) has been developed at FNAL to assess the suitability of materials for use in a large liquid argon time projection chamber. During development of the MTS, it was noted that controlling the cryostat pressure with a 'raining' condenser reduced the electron drift lifetime in the liquid argon. The effect of condensing has been investigated using a series of passive materials to filter the condensate. We report the results of these studies and of tests on different candidate materials for detector construction. The inferred reduction of electron drift lifetime by water concentrations in the parts per trillion is of particular interest.

Introduction

Liquid argon time projection chambers (LArTPCs) offer an opportunity for novel neutrino physics (1). They can provide bubble-chamber quality event images by drifting ionization electrons created by the passage of charged particles through the liquid to readout planes. Since argon is cheap and plentiful, one can conceive of detectors with mulit-kiloton active volumes. A principal challenge for large LArTPCs is the removal of electronegative impurities that capture the ionization electrons. The Material Test System (MTS) has been built at FNAL to develop liquid argon purification techniques (2) and measure the effects of materials on electron drift lifetime and qualify them for use in a large LArTPC. A schematic of the MTS cryostat is shown as Figure 1.

Some of the features of the MTS relevant to this note are the following.

- *Argon Source*. Commercial argon [] is passed through a molecular sieve (3) to remove water and activated copper (4) to remove oxygen and other electronegative impurities before entering the cryostat. The source provides liquid argon with a drift lifetime of many milliseconds [TM 2385]
- *Scrubber filter.* This wholly-internal filter contains a combination of molecular sieve (3) and activated copper (4). It can be used to maintain the purity of the argon in the cryostat and also to remove impurities that may be introduced during materials testing. A description of filter operation can be found in (2).
- *Lifetime monitor.* Modeled after the 'purity' monitors of the ICARUS Collaboration (make this Carugno reference and TM-2385), this device allows for the direct measurement of the electron drift lifetime.
- *Condenser to control cryostat pressure.* Argon vapor enters the condenser through a central tube and contacts surfaces cooled with liquid nitrogen pressurized to 50 psia to avoid freezing argon on the contact surface. The condensed argon flows down the condenser walls and drips into one of four condenser return paths before entering the bulk liquid. When the condenser is not operating, argon is continuously vented. A closed system is

desirable during materials testing so that material-introduced impurities remain in the cryostat and their effect on electron drift lifetime can be observed.

- *Return of condensed argon.* A wheel below the condenser allows the selection of a return paths for the condensate. There were four paths available: a 1.5 inch diameter tube with stainless steel wool enclosed in sintered metal, a similar tube with a disk of sintered glass, a thin spiral tube , and a hole which allows the condensate to fall directly into the bulk liquid. Fig. 3 shows details of this system.
- *Mechanism for material insertion.* An airlock separated from the cryostat by a large gate-valve sits above the cryostat. Materials may be placed into a sample cage inside the airlock and purged with argon vapor from the MTS cryostat. The gate valve is opened and the cage lowered into the cryostat using a plunger attached to the top of the cage. Once in the cryostat, the cage is set on a lift platform equipped with and a RTD to indicate temperature. The plunger can then be retracted, the airlock closed, and the material lowered into the liquid argon. Since it may not be possible to evacuate the cryostat of a future large LAr TPC, the MTS airlock is specifically designed to prepare materials for insertion by purging since evacuation might remove contaminants that otherwise would not be removed.
- *Data Acquisition Systems*. The lifetime measurement data-acquisition system consists of a Visual Basic program running on a Tektronix 5054NV digitizing oscilloscope. The system is fully automated and takes measurements at intervals set by the user. The cryogenics controls of the MTS are based on a Beckhoff controller with an Ifix interface. A communication program sends the lifetime data to the Ifix interface where it is stored with the cryogenic system information.

Operation of the MTS involves evacuating the cryostat, filling it with filtered commercial argon, inserting a material sample, and monitoring the electron drift lifetime. Upon evaluation, the sample may be removed and another sample material inserted. The condenser and internal filter may be operated as desired or needed.

Effect of Condenser Operation on Electron Drift Lifetime

After the initial success at obtaining long drift lifetimes at FNAL [TM-2385], the condenser was first used to control the MTS cryostat pressure in January 2008. The condensate was allowed to drip directly into the bulk liquid and it immediately became apparent that condensing reduced the electron drift lifetime dramatically, from ten milliseconds to less than 1 millisecond, as shown in Figure 2. [reference to ICARUS paper]

Since there was little material in the vapor of the cryostat other than the three coaxial lifetime monitor cables and the cryostat had been evacuated to below 10^{-6} torr, we did not consider that we were introducing significant chemical impurities by condensing the vapor and our first hypothesis was that the decrease in lifetime was due to the formation of Argon ions as the condensate dripped from the metal surface of the condenser and splashed onto the liquid surface. Our conclusion that this was not a chemical contamination was supported by the fact that operation of the internal scrubber filter improved the lifetime only a factor of 2, whereas we had been able to use the same filter to recover lifetimes ruined by deliberate injection of oxygen back to ten milliseconds.

Modifying the condenser would have been difficult since the liquid return is on the outside and the gas input is on the inside. We therefore installed a pipe under the condenser output with a section filled with stainless-steel wool enclosed with sintered metal discs, the idea being to offer any ion a nearby electrical surface. This addition to the system allowed us to obtain drift-lifetimes of several milliseconds. There was, however, still some uncertainty in our minds about the action of this pipe and,

to confirm the ion hypothesis, we replaced the steel-wool and sintered metal section of the return pipe with a section containing an electrically isolated metal rod at its center. Under a potential difference of 1.5 kV between the rod and the pipe, any argon ion would have plenty of time to reach an electrode given the flow rate of the condensate.

In practice we saw very little difference in lifetime whether the ion-rod was set to be a cathode or an anode or there was no potential applied, implying that the effect of the steel-wool and the sintered metal was not from discharging ions. When we examined the steel wool under a microscope to see if the effect was from trapping some particulate, the material was pristine – suggesting that if the metal was trapping something, the trapped material had evaporated when it was warmed to room temperature.

In an attempt to characterize the effect of condenser operation, a mechanism was installed beneath the outlet of the condenser that allowed one of four return paths for condensate return. The device is shown in Figure 1 and detailed in Figure 3. A return path is selected by rotating a handwheel that extends outside the cryostat.

The different return paths were chosen for their ability to differentiate between possible condenser-associated impurities, thought to be either ions or particulate. The thin, spiraled tube was designed to prevent the condensed argon from dripping into the bulk liquid. This would help prevent charge separation that results from fluid flow against a dissimilar surface (7) and prevent the generation of ions that might adversely affect the drift lifetime. The sintered glass was chosen for its ability to remove particulate, but not discharge any ions generated by dripping condensate. The sintered metal and steel wool were chosen for their ability to remove both ions and particulate. The length of the sintered metal and steel wool pipe was chosen to allow the end to be uncovered if the argon depth in the cryostat was below 18 inches – thus forcing the argon once again to drip out of a metal surface and splash onto a liquid surface. The hole provided a baseline to which to compare the effects of the other return filters.

The cryostat was initially filled with 29 out of 40 inches of argon, enough to cover the outlets of all the return filters except the hole. The effect of filtering the condensate through each of the returns was observed and results are shown in Figure 4. In order to better understand the effects of the return paths and scrubber filter operation, the impurity concentration in the cryostat was modeled assuming there were three "types" of impurities.

- [I1], a constant, base impurity concentration that limits the maximum electron lifetime.
  $[I1] = Base\ Impurities$
- [I2], the concentration of condenser-independent impurities. These impurities enter the liquid as they outgas from surfaces in the cryostat. Condenser-independent impurities accumulate in the liquid without active filtration.
  $$\frac{d[I2]}{dt} = (Outgassing\ Constant) * t^{-1/2} - (Active\ Removal\ Constant) * [I2]$$
- [I3], the concentration of condenser-associated impurities. These impurities are introduced into the liquid during condenser operation. It was assumed there was an infinite reservoir of condenser-associated impurities and that condenser operation introduced these impurities to the bulk liquid at a rate proportional to the heat load of the cryostat. Each of the return filters was assumed to reduce the rate of introduction by a constant fraction. Scrubber filter operation removes condenser-associated impurities; without active filtration these impurities still have an exit mechanism.
  $$\frac{d[I3]}{dt} = (Heat\ Load\ Factor) * (1 - Fraction\ Removed\ by\ Return) - (Active\ Removal\ Constant + Passive\ Removal\ Constant) * [I3]$$

The sum of these three impurity concentrations gives the impurity concentration in the liquid. This determines the electron drift lifetime. The unknown parameters of the models were found using a least-squares fit to the observed electron drift lifetimes. Table 1 gives these parameters.

Constant		Value	Units	Comment
Base Impurities		0.01		Determined from electron lifetime while venting with scrubber filter on.
Outgassing Constant		0.004	1/hour	Determined from fit.
Fraction Removed by Return	Hole	0		Defined as zero.
	Thin Tube	0.28		Determined from fit.
	Sintered Glass	0.58		Determined from fit.
	Sintered Metal	0.92		Determined from fit.
Active Removal Constant		0.63	1/hour	Determined from scrubber filter speed
Heat Load Factor		0.025 or 0.1	1/hour	0.025 if scrubber filter is off, 0.1 if on. Ratio determined by condenser LN2 consumption.
Passive Removal Constant		0.16	1/hour	Determined from fit

Table 1: Constants for Modeled Impurity Concentration in MTS Cryostat. The electron drift lifetime in milliseconds equals 0.15/([I1]+[I2]+[I3]). Some constants were estimated from operational parameters; others were determined from a least-squares fit to the observed electron lifetime.

The sintered metal and steel wool return path successfully removed a large portion (92 percent) of the condenser-associated impurities. However, performance of the other returns does not conclusively support the hypothesis that condensing introduces ions or ice/particulate into the bulk liquid. A new hypothesis was formed that condenser-associated impurities are emitted from warm metal surfaces but adsorb to cold metal surfaces. Return filter performance is related to the amount of cold metal surface area presented to the condensate. This hypothesis was adopted because it accounts for the difference in return filter performance as well as the passive removal of condenser-associated impurities from the bulk liquid. As a check on this hypothesis, the amount of cold metal surface area presented by the return paths to the condensate was decreased by lowering the liquid level in the cryostat. The return paths removed fewer condenser-associated impurities in this new operating condition, as shown in Table 2. The continued success of the sintered metal and steel wool return also eliminates the possibility of ions as the condenser-associated impurity. Ions would have been generated as the condensate dripped from the sintered metal return into the bulk liquid and decreased the electron drift lifetime.

Return Filter	Cold Metal Surface Area Presented to Condensate		Electron Lifetime	
	29 Inches LAr	*16 Inches LAr*	*29 Inches LAr*	*16 Inches LAr*
Hole	0	0	1.1	1
Thin Tube	150 cm^2	70 cm^2	1.5	1.3
Sintered Glass	300 cm^2	Near 0 cm^2	2.4	1.2
Sintered Metal	A lot	A lot	5 to 8	5 to 8
N/A (Venting)	N/A	N/A	10-20	10-20

Table 2: Electron Drift Lifetime as Related to Return Filter and Liquid Level.

Condenser-associated impurities can be actively removed with the scrubber filter, suggesting the source of these impurities is not liquid argon but the cryostat, which is evacuated before fill. Water

is well known to remain on metal surfaces in vacuum (8) and may be cryopumped, making it a likely appellation for condenser-associated impurities.

Material Tests and Inferred Effects of Water on Electron Drift Lifetime

A Tiger Optics moisture analyzer (9) with a 2 ppb detection limit and 1 ppb resolution was used to monitor the water concentration in the MTS cryostat. The argon vapor was monitored for moisture content because the moisture analyzer was not sensitive to concentrations in the liquid. When using the sintered metal return and operating the scrubber filter, we estimate the water concentration in the liquid is 1/500th of that in the vapor. This ratio varies depending on the operational parameters of the cryostat and associated apparatus and on the condition of the sintered metal return filter.

The airlock volume was connected to the cryostat volume to test the hypothesis that warm metal surface area introduces water and reduces the electron lifetime. During this test, the scrubber filter was operational and the sintered metal was used as the condenser return. The results, shown in Figure 5, provide some confirmation of our hypothesis. The water concentration in the argon vapor increases when the airlock is joined to the cryostat and also is an indicator of electron lifetime: the product of the electron lifetime and water concentration remains roughly constant.

A series of material tests was performed to determine the effect of various materials on the electron lifetime (see Table 2) and the role of water. Test materials were inserted in the airlock and evacuated and purged with argon from the cryostat and then lowered into liquid argon and subsequently raised into vapor. Material tests were conducted with the scrubber filter operational and the sintered metal as the return filter. During materials testing, it was noted that the water concentration in the argon vapor was indicative of electron drift lifetime (e.g. Fig. 6) in a way similar to that of joining the cryostat and airlock volumes: The product of the electron lifetime and the water concentration remained constant. In both scenarios, [Electron Lifetime in ms]*[H2O Concentration in ppb]≈17. It was also noted that upon evacuation in the airlock (for a few days) prior to testing, PC board materials had little effect on the electron drift lifetime (see Fig. 7). These characteristics suggest water may be the sole significant electronegative impurity introduced by various materials and metal surfaces.

Material	Sample Surface Area	Effect of Material on Electron Drift Lifetime (LT)			Comments
		94 K liquid	≈120 K vapor	≈225 K vapor	
GC Electronics Red-X Corona Dope on stainless steel	100 cm^2	None	None	LT reduced from 8 to 1 ms; recovery observed.	H2O Concentration not monitored. Sample has tendency to flake off substrate when cooled.
Deactivated Rosin Flux on stainless steel	200 cm^2	None	Not Tested	LT reduced from 8 to 1.5 ms; recovery observed.	H2O Concentration not monitored
FR4	1000 cm^2	None	Not Tested	LT reduced from 8 to <1 ms	Outgassed enough water vapor at 200K to saturate sintered metal return filter.
Taconic, Grade TPG-30	600 cm^2	None	Not Tested	LT reduced.	Sample outgases water at 225 K.
Hitachi BE 67G	300 cm^2	None	Not Tested	LT reduced; recovery observed	Sample outgases water at 225 K; outgassing reduced over time.
TacPreg	200 cm^2	None	None	LT reduced; recovery observed	Sample outgases water at 225 K; outgassing reduced over time.
FR4 board prepared by Brookhaven Nat.	225 cm^2	None	None	LT reduced from 8 to 3 ms.	Sample is a y-plane wire endpoint for MicroBooNE detector. Sample outgases water at 225 K.

Lab.					
FR4 board prepared by Brookhaven Nat. Lab.	225 cm^2	None	None	None	Sample is a y-plane wire endpoint cover for MicroBooNE detector. Sample was evacuated before introduction to cryostat.
Devcon 5-minute epoxy	100 cm^2	None	None	LT reduced from 10 ms to 6 ms; some recovery observed	Sample outgases water at 225 K.

Table 2: Summary of Material Test Results. "LT" refers to the electron lifetime. Most materials began outgassing water and reduced the electron lifetime when raised to 225 K. When maintained at this temperature for several days, this outgassing decreased for some materials and there was a corresponding increase in the electron lifetime. The water concentration of the argon vapor was not monitor for the first two material tests for lack of instrumentation. In all of the material tests in which the water concentration was monitored, it was a good indicator of the electron lifetime.

While materials testing and characterization are incomplete, the results presented in Table 2 indicate a variety of materials outgas water when warm (≈225 K) and affect the electron lifetime in the MTS cryostat. This outgassing has been observed to initially decrease over a period of a few days in certain materials; the extent of its decrease has not been characterized.

Summary and Conclusions

Investigation of the relationship between raining condenser operation and electron lifetime in the MTS cryostat and the results of material tests indicate that low (10 ppt) concentrations of water in liquid argon affect the electron drift lifetime. We have not demonstrated a direct relationship between water concentration in liquid argon and electron drift lifetime. However, based on our results, we suggest water moves through the cryostat in the following way. Warm metal surfaces and other unevacuated, warm, and (perhaps) recently-introduced materials release water into the argon vapor. Condenser operation introduces the water-contaminated vapor into the liquid of the cryostat where water naturally exits the liquid because of its affinity for cold metal surfaces. The equilibrium concentration of water determines the electron lifetime. Water may be prevented from entering the liquid by filtering the condensate before it enters the liquid.

Figures

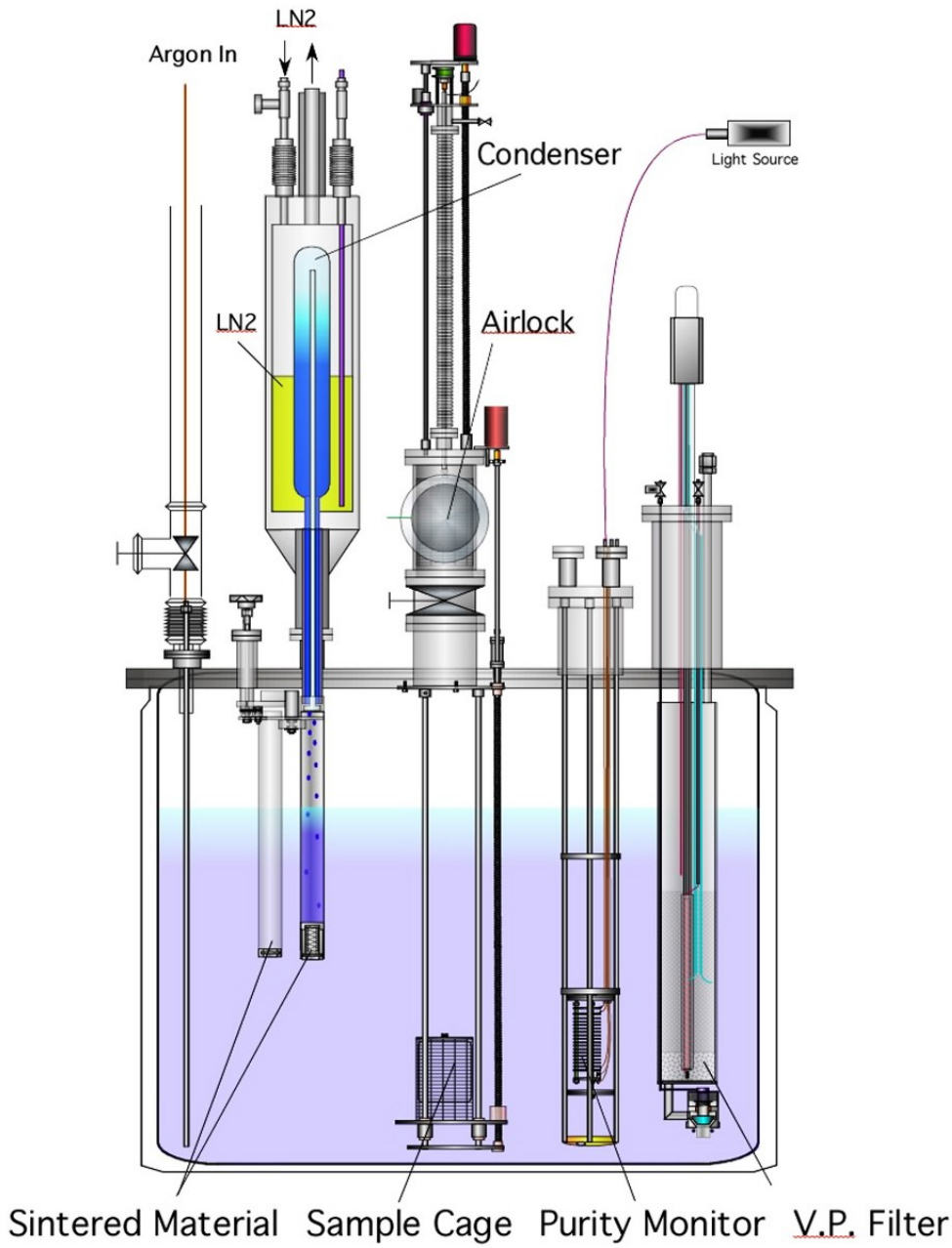

Figure 1: Schematic of the Materials Test System (MTS) cryostat at FNAL.

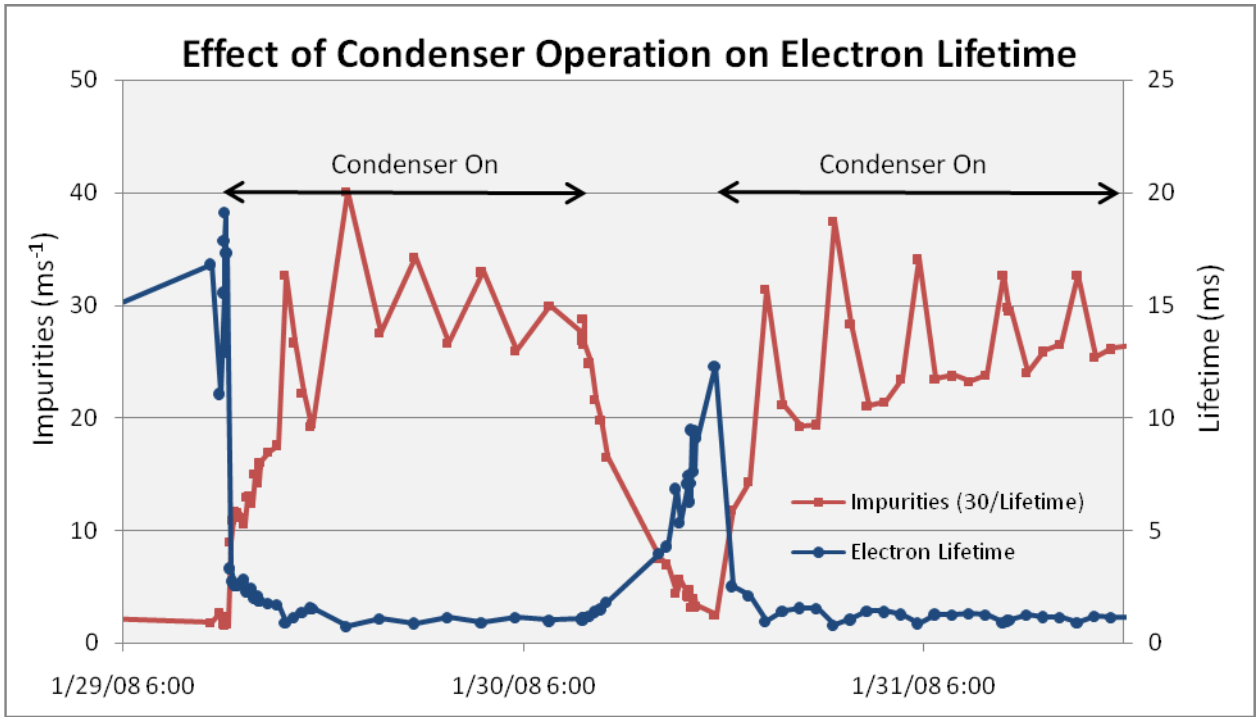

Figure 2: Effect of Condenser Operation on Electron Drift Lifetime. The impurities, her defined as 30/Lifetime, are useful because they represent the physical contaminants in the argon. When the condenser is off, the electron lifetime approaches 20 ms; when the condenser is on, the lifetime quickly degrades to 1 ms or less. The oscillations in the electron lifetime and impurities are related to cycling of the condenser.

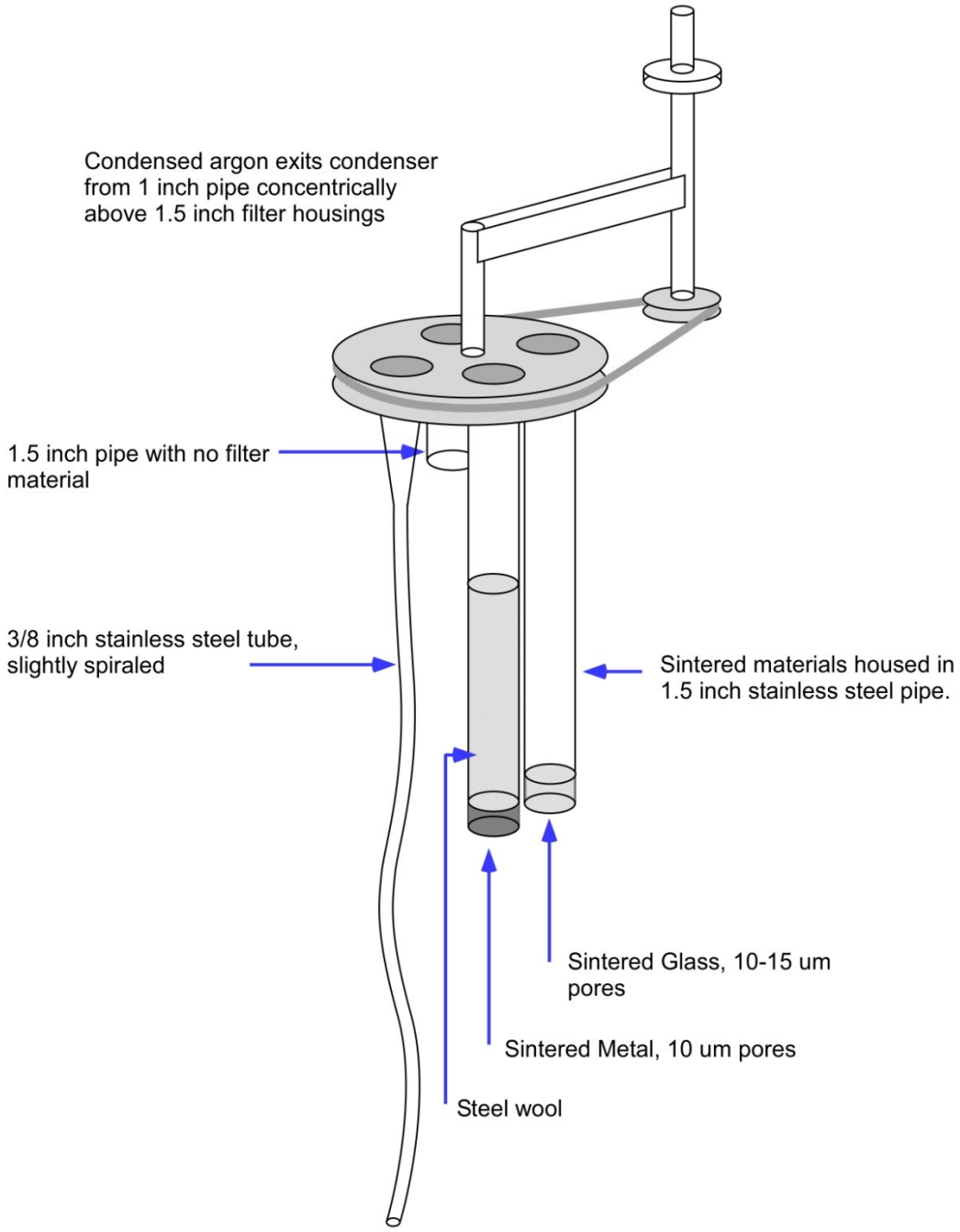

Figure 3: Detail of Return Filter Mechanism. The return mechanism contains four return paths: a thin tube, sintered glass, steel wool and sintered metal, and a pipe stub with no filter media. The thin tube extends approximately 36 inches into the cryostat, which has a depth of 40 inches. The housings for the sintered metal and sintered glass extend approximately 20 inches into the cryostat. A handwheel, fed through the top flange of the cryostat, is used to select the return path for the condenser.

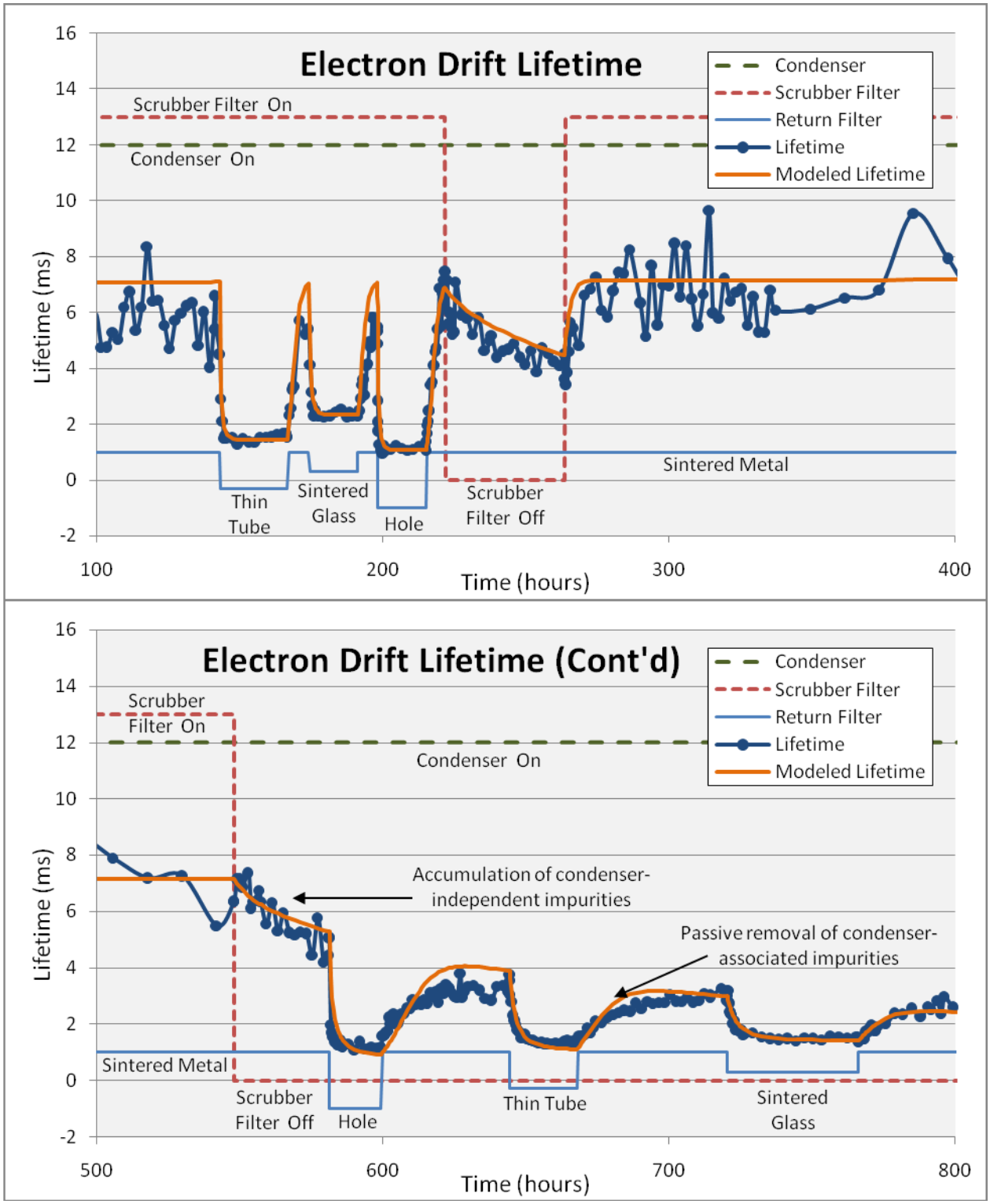

Figure 4: Electron Drift Lifetime as a Function of Condenser Returns and Scrubber Filter Operation. The dashed red line and the dashed green line indicate, respectively, scrubber filter and condenser operation: high for on, low for off. The light blue line and its associated labels indicate which condenser return is in use. The dark blue shows the observed lifetime and the orange line represents the modeled lifetime. The model is discussed in the text. The figure shows the effect of the different return filters on the electron lifetime with the scrubber filter both on

(first half of Fig. 4) and off (second half of Fig. 4). When the condenser is off, electron lifetime is between 10-20 ms.

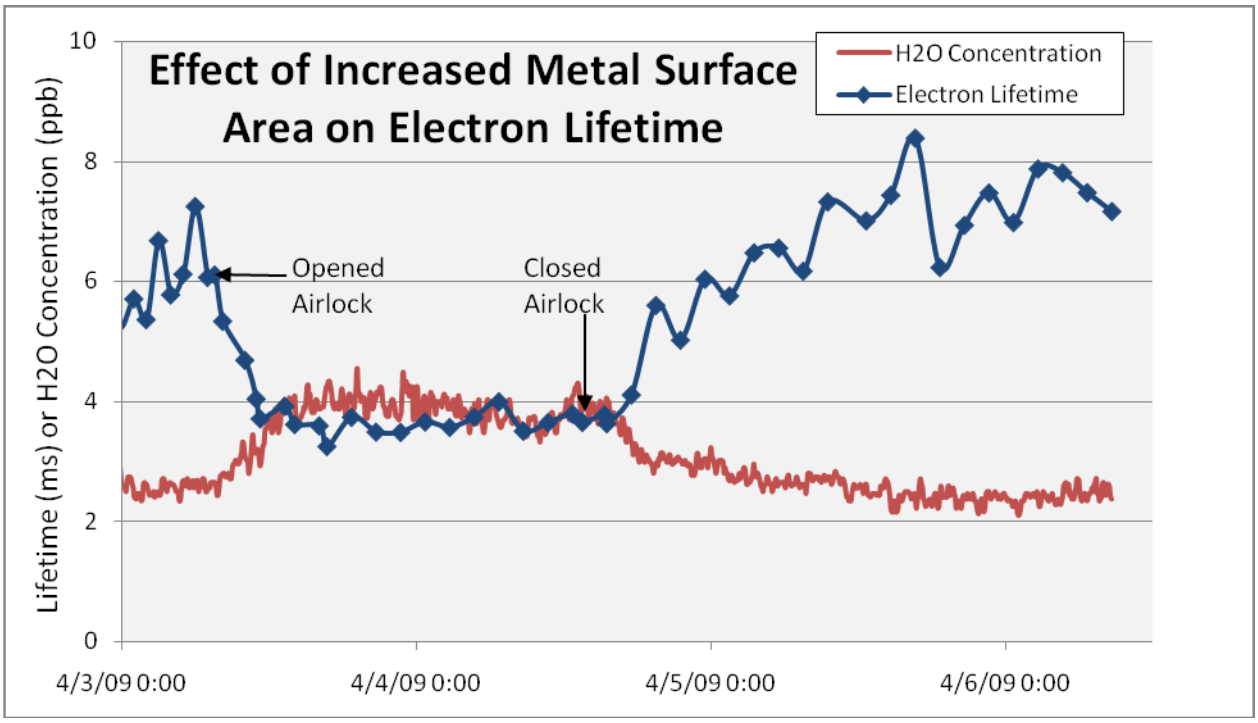

Figure 5: Effect of Connecting Cryostat and Airlock Volumes. The test was performed with 15 inches LAr present in the cryostat. The cryostat was connected to the airlock by opening the gate valve that typically separates the two. Prior to opening, the airlock was under vacuum. The results of opening the airlock are attributed to the additional metal surface area introduced to the argon vapor. The relationship between water concentration and electron lifetime is similar the relationship observed during materials tests (e.g. Figure 6).

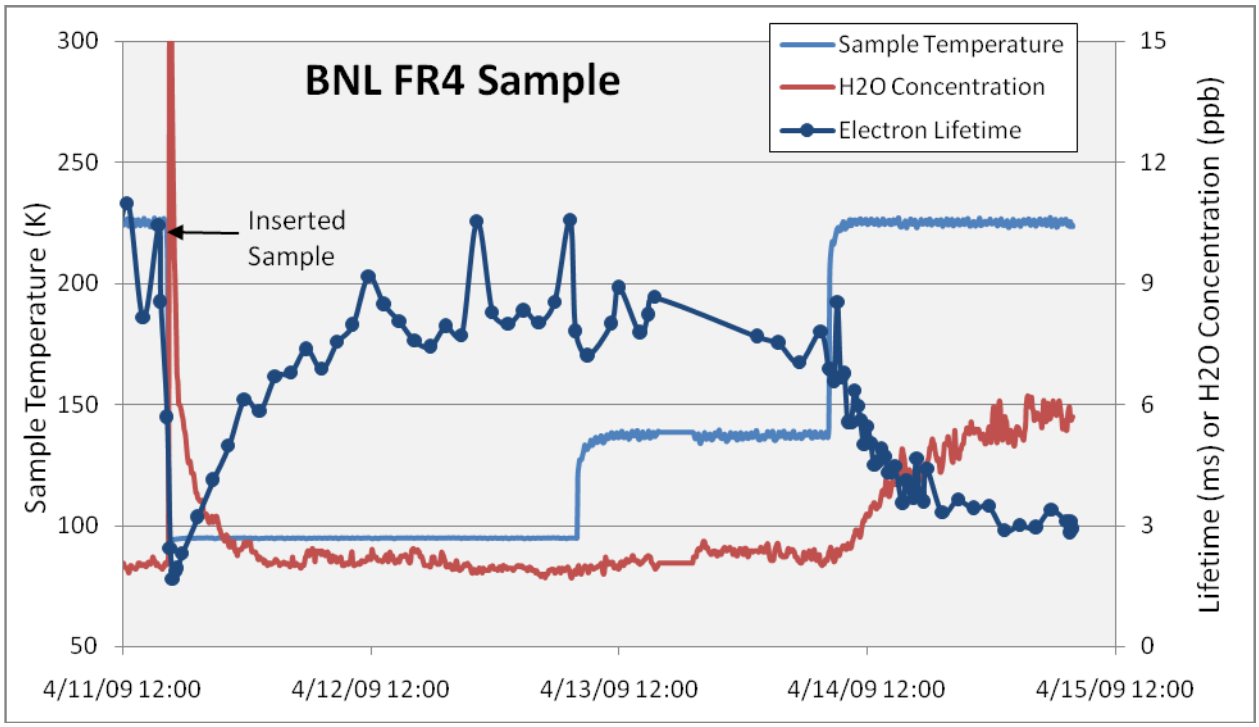

Figure 6: Material Test of BNL FR4 Y-Plane Wire Endpoint. The material test was performed with 17 inches LAr present in the cryostat. The sample was first lowered into the liquid argon then raised so that the temperature of the sample was increased. When moved to 225 K, the sample began to outgas; the effect on water concentration and electron lifetime can be seen in the figure. A similar relationship between water and lifetime was observed during other material tests, including the test documented in Figure 5.

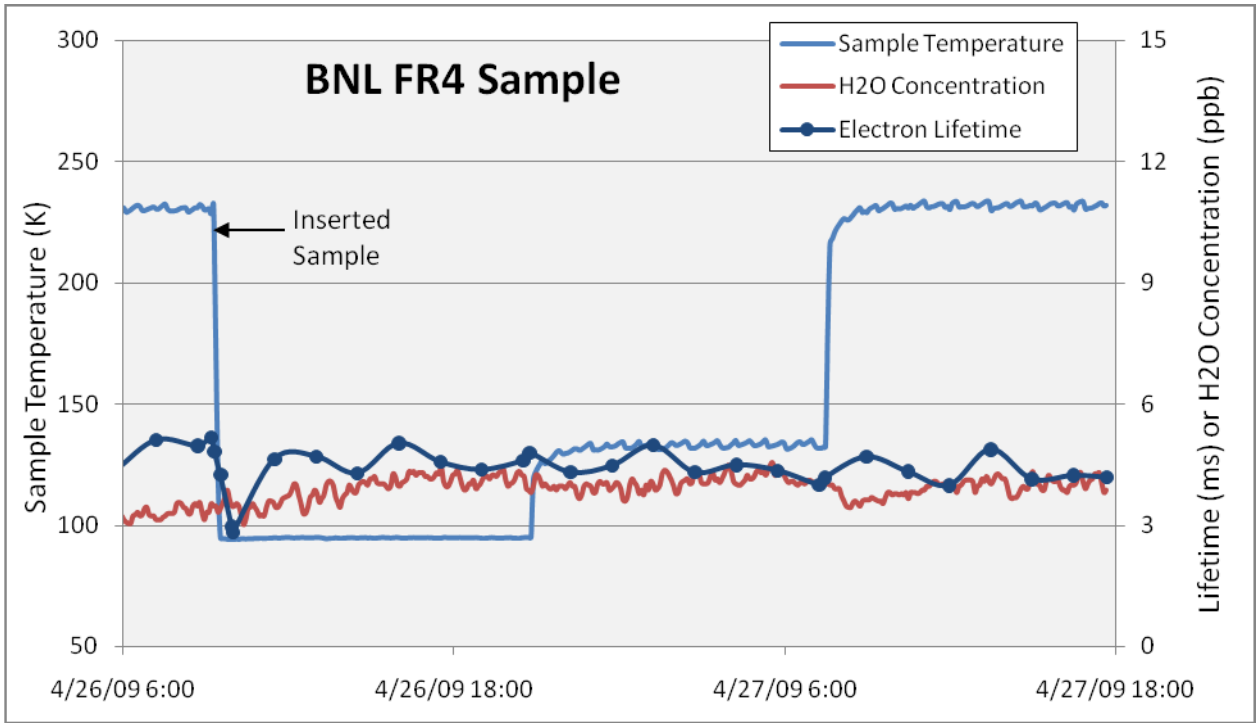

Figure 7: Material Test of BNL Y-Plane Wire Endpoint. The BNL FR4 sample was placed in the airlock and evacuated to 1 mTorr for a few days before being lowered into the liquid. When moved to 225 K, the sample did not outgas any water and had no effect on the electron lifetime. The material test was performed with 13 inches LAr present in the cryostat.

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