Purity Analysis For Xenon Detectors

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1 Introduction

Liquid xenon is commonly used as a detection medium for modern particle physics experiments, such as neutrino mass and WIMP dark-matter searches. Xenon is a good detection medium having high scintillation yield and, being a noble element, charge can be drifted and collected with relative ease. Achieving necessary purity for large-scale xenon detectors is non-trivial due to out-gassing of detector components such as Teflon, commonly used as a reflector. To drift electrons over tens of centimeters the xenon should contain no more than 1ppb (part per billion) equivalent of O_2 [1]. Dark matter detectors also have stringent limits on the krypton content of the xenon requiring a residual krypton concentration of than 1ppt (part per trillion) [2,3,4,5,6]. For a ton-scale xenon detector, just one standard liter of air (1 g) contains enough krypton to compromise background goals for a WIMP search experiment. Air leaks are of great concern since all experiments rely on recirculation-pumps for purification, and are prone to air-leaks. Purity monitoring using electron lifetime in liquid xenon is implemented in all experiments but has an Achilles-heel. Recirculation pumps, being the dominant source of air leaks in a recirculation system, are always followed by purifiers intended for removing N_2 and O_2. If an air leak is to occur then the electronegative impurities will be continuously removed so the electron lifetime remains unaffected, while argon and krypton would pass through the getter and accumulate in the bulk xenon. Relying on electron lifetime measurements alone will be insufficient for next generation ton-scale detector, a purity monitoring program of gaseous xenon will be necessary.

My work in the last two years have focused mainly on gas phase xenon purity analysis. I have helped to develop and implement a purity analysis system in EXO-200 and LUX. Using the coldtrap mass spectroscopy technique we are able to identify individual problematic species in xenon such as N_2, O_2, CH_4, Ar, Kr, He and measure them to sup ppb levels. In the case of krypton we have demonstrated sensitivities to less than 1ppt in the lab, already sufficient for future generation dark-matter detectors. Monitoring gas phase purity at such low concentrations has allowed us to begin studying how impurities interact between the liquid-gas interface, which is not well understood for the case of xenon.

2 Double Beta Decay

The EXO-200 experiment is a neutrino mass search using 200 kg of xenon enriched to 80% of the isotope ^{136}Xe. Many isotopes undergo double beta decay, however the beta decay mode is typically dominant and swamps the double beta decay signal [7]. ^{136}Xe is one of a handful of isotopes in which the single beta decay mode is forbidden, it only double beta decays to ^{136}Ba. This allows for a unique ability to test for the majarona nature of the neutrino and to probe the neutrino mass.

In a standard double-beta decay two neutrons decay to two protons, electrons and anti-neutrinos. Since the two neutrinos carry away some of the energy of the decay and escape detection a typical beta decay spectrum is observed. If neutrinos are majorana particles then it is possible for the two neutrinos to annihilate in the double beta decay process. In this special case, the two electrons carry away the energy of the decay (Q=2.6Mev) [8]. Thus, an excess of events at the endpoint of the double beta decay spectrum is evidence for neutrino-less double beta decay. Figure 1 shows an example of a double-beta decay spectrum with a magnified view of the neutrino-less double-beta decays at the end of the spectrum. The half-life of this rare process can than be related to the inverse square of the absolute neutrino mass [9].

Neutrinos are known to have mass from oscillation experiments[10,11,12,13]. The mass difference of solar neutrinos are found to be 50meV and the mass difference of atmospheric neutrinos are know to be 5meV. If an experiment can reach sensitivity of less than 5meV then the inverted hierarchy nature of the neutrino can be

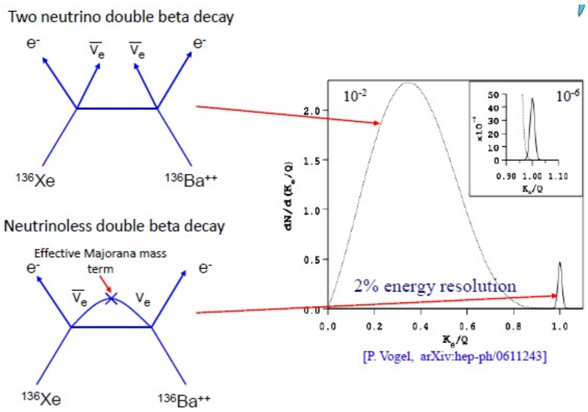


Figure 1: Top Left: Standard double beta decay, resulting in two electron and two neutrinos in the final state. Bottom Left: Neutrino-less double-beta decay with only two electrons in the final state. Right: Double beta decay spectrum with the predicted excess of events at the endpoint from neutrino-less double-beta decay.

ruled out, see Figure 2. In order to set such a strenuous limit it would require Full-EXO, a 10Ton of enriched xenon running for ten years.

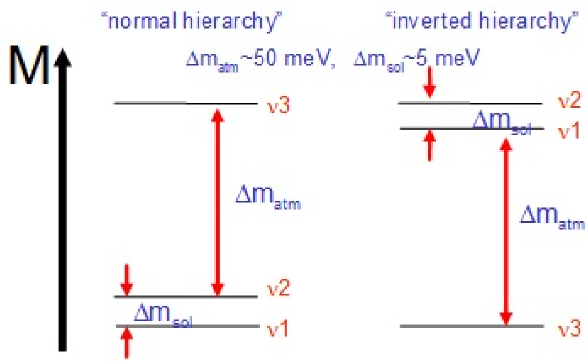


Figure 2: Left: Normal neutrino mass hierarchy. Right: Inverted neutrino mass hierarchy.

3 Weakly Interacting Dark Matter

Astronomical observations predict the existence of non-luminous matter, dark-matter. Evidence for dark-matter include galactic ration curves, gravitational lensing anomalies. At large distances the velocity of objects around the center of a galaxy does not fall off as predicted by Newtonian physics. Instead the velocity distributions appear constant as if a halo of 'non-luminous mass was surrounding the galaxy. The rotation curve of our own Milky-way galaxy requires the presence of dark-matter [14]. Astronomical observations of the bullet-cluster also support the existence of dark-matter. The bullet-cluster is made up of two galaxy clusters which have recently collided and passed through each other. The collision has caused the ordinary matter to heat and emit xrays, from elector-magnetic interactions, allowing for the mass distribution to be mapped [15]. However, the observed concentration of mass is not consistent with the center of mass predicted by GR, specifically gravitational lensing [16]. The way light bends around the bullet-cluster would indicate the presence of a dark-matter shell which, unlike the ordinary matter, has passed through at a faster rate due to the lack of interactions. Figure 3 shows the concentration of mass in the bullet-cluster as observed from xrays, emitted by ordinary matter, in pink and the concentration of mass from gravitational lensing in blue.

Figure 3: Bullet Cluster. The pink represents the mass distribution in the bullet cluster from x-rays observed by Chandra. The blue represents the mass distribution in the cluster as mapped my gravitational lensing. [15,16]

LUX is a direct detection dark-matter experiment using 350 kg of liquid xenon. Recent theoretical dark-matter models favor the existence of WIMPs (weakly interacting massive particles). These WIMPS could have masses in the GeV to TeV range and would interact via the weak force with ordinary matter. The distribution of WIMPs in our galaxy is estimated from galactic rotation curve of the Milky-Way. The local density of WIMPs near the earth, at 8 kpc from the galactic center, is about 0.3GeV/cm^3, see Figure 4 [ref]. Given an assumed WIMP mass of 100GeV this corresponds to only about three proton masses per liter of space. The velocity of WIMPs near the Earth is about 240km/s (see Figure 5) [14]. WIMPs being highly non-relativistic would scatter coherently off of target nuclei with a cross-section corresponding to $\sim \mathrm{A}^2$. Xenon being a relatively heavy element, A=131, makes it an ideal candidate for a WIMP dark-matter search. Other common detection mediums are germanium, which is rather expensive on the ton scale, and argon which is cheap but suffers in cross-section and contains a troublesome radioactive isotope ^{39}Ar. Given that WIMPs are so sparse and interact rarely with ordinary matter a ton scale xenon experiment may only see a handful of events per year, thus having low backgrounds is crucial.

When a WIMP interacts with the target xenon it will, in most cases, cause a nuclear recoil event. The energy deposited by WIMPs is on the order of several keV. LUX can reject most, but not all common high energy backgrounds emanating from detector components, such as K,CO60,U,Th, because the gammas they emit are high energy ($>$ 1MeV). Low energy external backgrounds which could mimic are WIMP signals are greatly suppressed by the self-shielding of liquid xenon, see Figure 8. Electronic recoil backgrounds are further reduced by about a factor of two-hundred by comparing scintillation to ionization energy of events, nuclear recoil events tend to cause more scintillation than ionization in liquid xenon (Figure 6). Neutrons also cause nuclear recoil events in the detector, however they are likely to multi-scatter in the xenon and are greatly suppressed with an active water shield.

Without additional processing of the bulk xenon the dominant background for a xenon based dark-matter experiment turns out to be low-energy radioactive isotopes contained in the xenon its-self, see Figure 7. Xenon is distilled from the air and typically contains argon and krypton at ppb to ppm levels. Argon contains ^{39}Ar with an isotopic abundance of 10^{-16}, a half-life of 269years and a Q value of 565keV [17]. Even worse is ^{85}Kr, a beta emitter with an isotopic abundance of about 2e-11 (mol/mol), a half-life of 10.8 years and a Q value of 687keV [18]. Next generation dark-matter experiments will require that the residual krypton content of the xenon be less than 3ppt to probe cross-sections of 10^{-47}cm^2[3,4,5]. Residual krypton, and argon, can be removed via

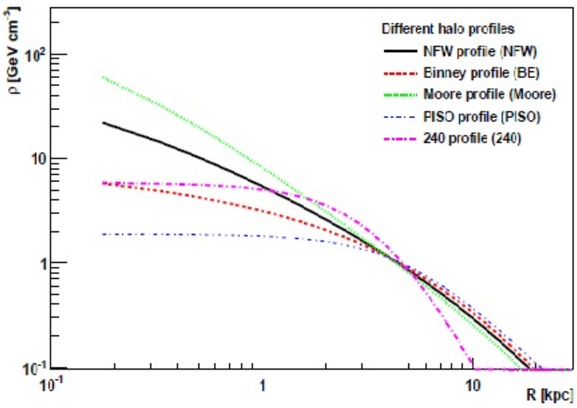
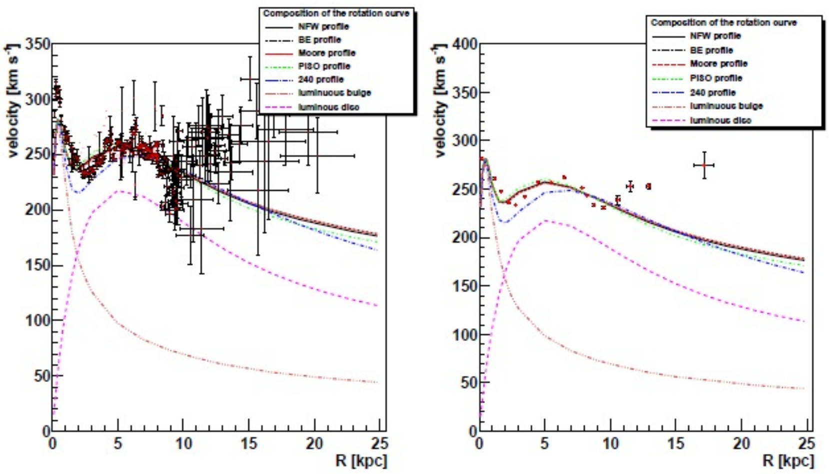


Figure 4: Dark-matter density vs. distance from the galactic center for several halo profiles of the Milky-Way.



(a) Rotation curve ($z = 0$ kpc), non-averaged

(b) Rotation curve ($z = 0$ kpc), averaged

Figure 5: Left: A plot of the rotational velocity vs. distance from the galactic center of the Milky-Way. Right, the average of measured rotational velocity vs. distance from the center of the Milky-Way.

gas chromatography or distillation-columns yet measuring the residual krypton content after processing is rather difficult.

4 Radioactive Background Requirements

LUX has several key advantages for background requirements over EXO. LUX will be looking for low energy nuclear recoil events ($\sim$ 10keV) and can simply reject most naturally occurring radioactivity with an energy cut. Only rare forward scatting events would deposit energy in the 10keV range. Low energy external backgrounds also pose little concern as they do not travel far in liquid xenon and can be rejected by taking a fiducial cut [see Figure 8]. Additionally, LUX searches specifically for nuclear recoil events and can reject one in every two-hundred electronic recoil events. Even with all the benefits from background rejection, high energy gammas from PMTs

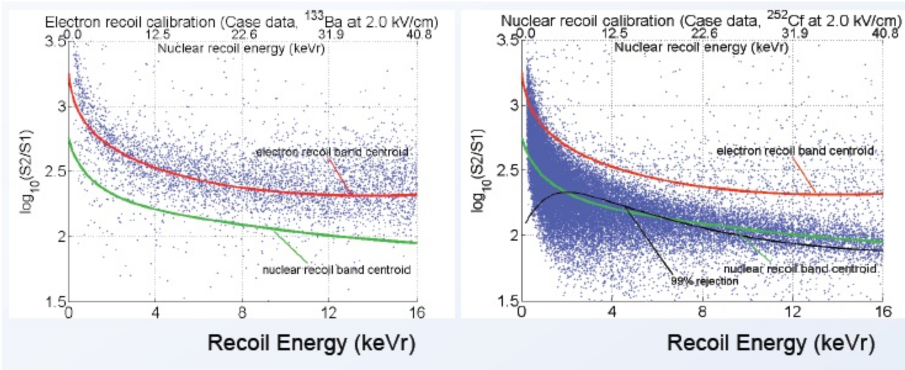


Figure 6: Left: The scintillation to ionization ratio of an electron recoil calibration in liquid xenon. Right: The scintillation to ionization ratio of an nuclear-recoil calibration in liquid xenon.

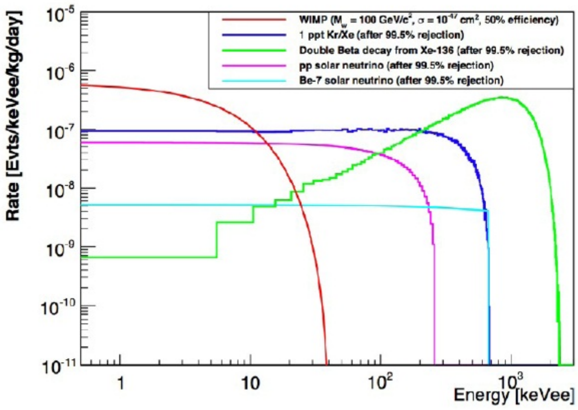


Figure 7: Expected event rate in a ton-scale liquid xenon detector. Assuming a 100 GeV wimp mass and 1ppt of residual krypton contamination.

are the largest background after krypton removal from the xenon. Krypton contains the radioactive isotope ^{85}Kr which is a low energy beta emitter and will be be dissolved uniformly in the liquid xenon its-self, this is especially troublesome because ^{85}Kr events occur within the fiducial volume. For example, if LUX meets its goal of 3ppt krypton and taking into account the isotopic abundance of ^{85}Kr, $\sim 2 \times 10^{-11}$, there will be about 80,000 ^{85}Kr atoms in the xenon. With a half-life of 10.8 years, the number of ^{85}Kr beta decays between 0-10keV in the fiducial volume per year comes to about 70, before applying the 1/200 electronic recoil rejection. Even a handful of events per year is a large background when searching for WIMPs with such tiny cross-section $< 10^{-46}$cm^2. A minor air leak could easily introduce 100ppt krypton into LUX mimicking several hundred WIMP like events per year.

EXO, on the other hand, searches for electronic recoil events at energies around 2.5 MeV and therefore does not benefit from any of the background rejection techniques that LUX uses. This means that the materials used in the EXO detector must be extremely pure of trace radioactivity such as K,CO60,U,Th. In the case of the regular two neutrino double beta decay EXO does get interference from it's residual krypton contamination. The turn around of the 2nBB spectrum for ^{136}Xe occurs at around 800keV which is close to the endpoint of ^{85}Kr at 687keV. EXO-200 has recently measured the half-life of ^{136}Xe to be 2.1×10^{21} years by using a lower energy cut at 700keV. To observe the turn around of the 2nBB spectrum the energy threshold will be lowered in later data analysis. Based on the measured half-life of ^{136}Xe about 80,000 additional events per year are expected. The krypton content of the xenon was measured by our cold-trap technique to be 25+/-3 ppt, this corresponds to 35,000 beta-decays per year that can not be rejected in the EXO-200 detector. Once the energy threshold is lowered, the krypton content will become a major background of the ^{136}Xe 2nBB spectrum for the EXO detector.

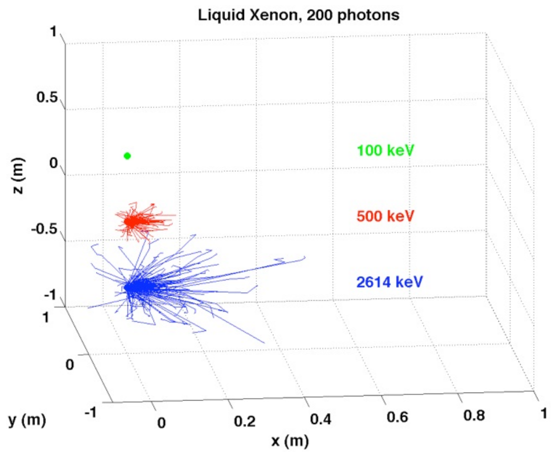


Figure 8: Monte Carlo simulation of gammas penetrating into liquid xenon.

5 Cold Trap Xenon Purity Analysis

5.1 Need For Xenon Gas Purity Measurements

All liquid xenon experiments require the removal of electronegative impurities, such as O_2, to be able to drift electrons in the liquid. Screening for O_2 and N_2 before filling of a detector can save experiments from depleting the purifiers used for electronegative impurity removal, a non trivial cost. Dark-matter experiments using liquid-xenon also require that the residual Kr concentration be at the ppt level after processing via distillation or gas-chromatography. An analytic method for monitoring gas-phase krypton purity of the xenon after processing would prove useful to confirm the effectiveness of the krypton removal process. The xenon used for LUX will be processed via gas chromatography which will potentially introduce levels of helium which could degrade the PMTs used in the experiment. It will be important to verify the helium content of the xenon is sufficiently low before filling the detector. PMTs are damaged when helium diffuses through the glass of the PMT degrading the vacuum, this leads to after-pulsing.

5.2 Coldtrap Purity Measurement Technique

We have developed a technique for observing small concentrations of impurities in xenon gas using a residual gas analyzer (RGA). The purity analysis technique involves sampling the xenon of interest with a ultra-high vacuum leak-valve, passing the xenon through a liquid nitrogen cold-trap, and sampling it with a residual gas analyzer, while the RGA is being pumped to vacuum with a turbo-molecular pump [see Figure 9]. The RGA requires a vacuum of better than 1e-5 Torr to operate and would be over-pressured from the bulk xenon without the coldtrap limiting impurity detection to about ppm levels. The coldtrap serves to remove the bulk xenon while allowing impurities of interest to pass through for analysis. This simple technique boosts the sensitivity of the RGA by at least five orders of magnitude to impurities of interest such as N_2, O_2, CH_4, He, Ar, Kr.

Once xenon ice has formed in the coldtrap the vapor pressure of xenon is held fixed regardless of the xenon mass at 1.8mTorr, the vapor pressure of xenon at 77K [19]. Many impurities of interest can pass through the

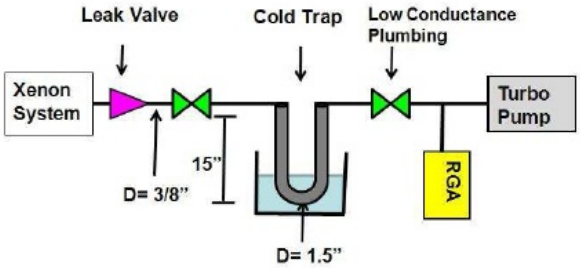


Figure 9: Coldtrap purity analysis system diagram.

coldtrap as the conditions in the coldtrap, 1.9m Torr at 77K, are below the liquid-vapor or solid-vapor equilibrium. With the xenon pressure held fixed the leak-valve can be opened further to allow for vastly higher leak rates through the RGA. The sensitivity of the RGA to an impurity's partial-pressures is directly proportional to the leak rate as more impurities pass through the RGA per unit time for analysis. Thus, the trick to the technique is that the bulk xenon is removed, maintaining vacuum in the RGA, while the flow of impurities through the RGA is maximized. Maximum sensitivity is achieved by using the maximum allowable flow-rate, which is ultimately limited by the argon content of the xenon. Even at the ppm level, argon will overtake the xenon ice vapor pressure and begin to saturate the RGA at a flow-rate of about 0.1SLPM.

The partial-pressures of impurities read by the RGA for a given leak rate can be calibrated in terms of absolute concentration by injecting purified xenon a known amount of impurities and analyzing it. It was found that the partial-pressure is proportional to the absolute concentration of the impurity normalized to the flow rate through the leak-valve [20]. We have demonstrated in previous studies that oxygen, nitrogen, methane and krypton pass through the cold-trap largely undisturbed for analysis with a partial-pressure proportional to absolute concentration. The sensitivity to oxygen, nitrogen, and methane was found to be 0.66×10^{-9}, 9.4×10^{-9}, and 0.49×10^{-9} (mol/mol), respectively [20]. Figure 10 shows the result of a typical purity measurement. At time T=0 the xenon ice had already been formed and the RGA backgrounds of the masses of interest had stabilized. At time T=10min the leak-valve is opened and xenon flows through the analysis system at about 0.1SLPM. The xenon partial-pressure signal in the RGA does not rise because the bulk xenon is being removed by the coldtrap. Impurities such as Argon, Kr, and N_2 give clear signals, the specific sample was purified of O_2. At T=28min the flow is cut off and the partial-pressure signals in the RGA begin to return to background levels.

5.3 Measuring Krypton to sub ppt Levels

While measuring impurity concentrations to the ppb level for most species is useful it is not sufficient for krypton in xenon. The krypton requirement for LUX is less than 3ppt and next generation experiments will require residual krypton concentrations of less than 1ppt. [3,4,5] We were able to extend the coldtrap technique to measure sub ppt concentrations of krypton by creating a highly purified sample of xenon and using faster leak-rates through the coldtrap analysis system.

Using a rather small 40g sample of de-kryptonated and de-argonated xenon the coldtrap analysis system was tested for krypton sensitivity. Removing the argon was necessary for achieving higher flowrates without saturating the RGA. Samples containing between 0.5ppt and 2ppb were prepared with known concentration of krypton and then measured, see Figure 11[21]. The response of the coldtrap analysis system to krypton was linear in concentration over four orders of magnitude. Even at sub ppt injection a clear krypton signal could be seen by the RGA [21]. The sensitivity was limited by the flowrate of 1.5 SLPM due to the rather small sample of xenon used. With a larger sample of de-kryptonated and de-argonated xenon we expect to trivially achieve ten times the flow rate and thus ten-times the krypton sensitivity. Even so, detection of krypton to the ppt level

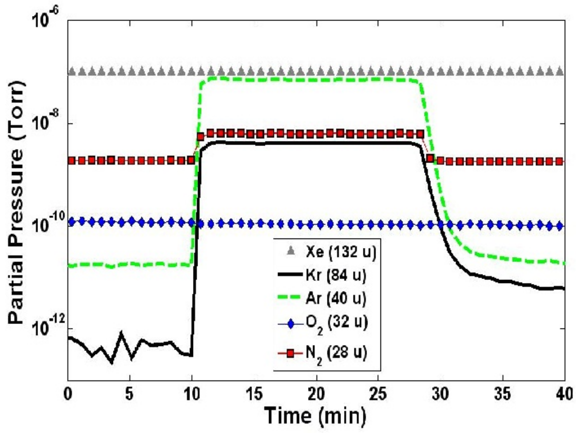


Figure 10: A typical coldtrap purity measurement. At T=0 xenon ice has already been form and signals have reached backgrounds. At time T=10 the leak valve is opened and xenon flows through at 0.1 SLPM. At T=28 the leak valve is closed.

will prove useful for the current and next generation of liquid xenon dark-matter experiments.

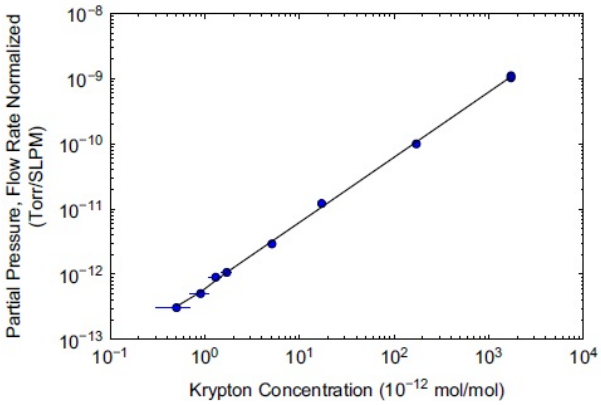


Figure 11: Results of krypton injections over four orders of magnitude. The response of the RGA is linear as a function of injected krypton concentration down to sub ppt levels.

6 Coldtrap for Understanding Liquid-Gas Purity

Having the ability to analyze specific impurities in the gas phase has yielded some unexpected results about solubility and the interactions of impurities between the gas and liquid phases in a detector. The first clue for solubility was from samples taken from the EXO-200 detector during the natural xenon run, see Table 1. The concentration of He, Ar, and Kr in the xenon were measured before liquefying the xenon, while the detector was full of liquid, and after liquid recovery. It was found that while liquid xenon was present in the EXO-200 detector the amount of He,Ar and Kr was much less than the bulk xenon average, even while flowing at a modest flowrate of 6 SLPM. The result was an indication that the impurities preferred to remain dissolved in the liquid xenon rather than be in the gas phase. This behavior is a potential issue because it makes the monitoring of the purity impossible via gas sampling while liquid is present in the detector.

ppb (g/g)	O_2	N_2	Ar	He	Kr
Natural Xe (after recovery)	<1	110 ± 25	255 ± 50	260 ± 80	43 ± 16
TPC full of liquid	<1	<10	1.0 ± 0.2	<0.04	4.4 ± 1.7
Xe Remaining after recovery	<1	<10	1900 ± 400	3100±1000	120 ± 50

Table 1: Purity results of the EXO-200 natural xenon run before, during and after xenon liquefaction.

A series of experiments using our xenon system at UMD were conducted to better understand the relationship between gas phase and liquid phase purity. A vessel with 3kg of liquid xenon was used to test the solubility of a specific species in xenon. The xenon was prepared with know amounts of impurities and measured with the coldtrap before liquefaction. Once the xenon was liquefied, the impurities in the xenon-gas above the liquid were remeasured. The results of such a test are show in Figure 12. It was found that when the xenon was left to circulate at a relatively small rate (0.2SLPM) the impurities would disappear from the gas phase, indicating that they dissolved and remained in the liquid. With the recirculation pump on at 5 SLPM however, the impurities in the gas phase appeared at an over abundance of about ten times. Apparently, by perturbing the liquid-vapor pressure equilibrium above the liquid impurities are ejected from the liquid, much like the release of CO_2 when a bottle of Cola is opened. The result suggests that if sufficient circulation can be reached in a liquid xenon system then impurities could be made to be readily ejected from the liquid.

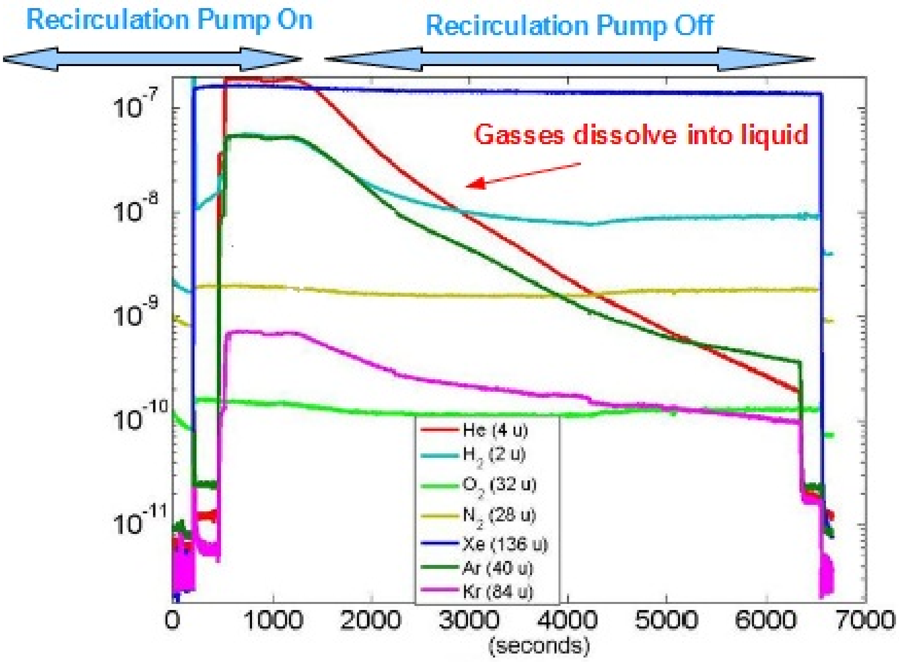


Figure 12: The reduction in He, Ar, Kr in gaseous xenon after the recirculation pump is turned off. The disappearance from the gas phase is attributed to adsorption into liquid xenon.

7 Henry's Constant Measurements

Measurements of Henry's constants with our set up at UMD were done for He, CH_4, N_2, O_2, Ar, and Kr. The Henry's constant is the measure of the concentration of impurity in the gas above the liquid (in mol/mol) divided by the concentration of impurity in the liquid its-self (in mol/mol). Thus, the larger the Henry's constant the less likely an impurity is to be dissolved in the liquid. The Henry's constants for these species where measured by preparing our 3kg of xenon with know concentrations of impurities and confirming the impurity concentration by measuring it with the coldtrap. Then the xenon was liquefied and the gas above the liquid was analyzed for the impurities remaining in the gas above the liquid.

For He, CH_4, and Kr we studied the Henry's constant as a function of liquid xenon temperature. The slope of the log of the Henry's constant is the enthalpy of dissolution, or the energy released when the impurity is dissolved or removed from the liquid. Figure 13 and Table 2 show the results found for He,CH_4 and Kr. The enthalpy of dissolution H is defined as $\mathrm{H} = -\mathrm{R} \times \frac{\mathrm{dln(K)}}{\mathrm{d(1/T)}}$. Where R is the gas constant, K is the measured Henry's constant as a function of temperature, and T is temperature.

	C(K)	H (KJ/mol)
Kr	3777	-31.4
CH_4	3145	-26.1
He	-19762	164

Table 2: The log of measured Henry's constants for He,CH_4,Kr vs inverse temperature.

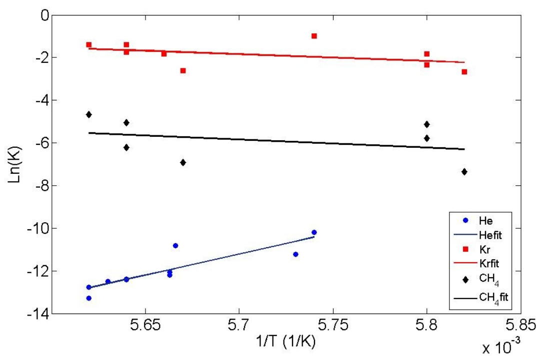


Figure 13: The log of measured Henry's constants for He,CH_4,Kr vs inverse temperature.

8 Conclusion

We have developed gas purity monitoring system which have proved useful for two large-scale xenon detectors. With the technique was can analyze specific species of impurities via mass-spec analysis. Monitoring gas phase purity has proved useful for ensuring that experiments can meet their purity goals, either for removing electronegative impurities or residual radioactive elements in the xenon its-self. By studying impurities on both EXO-200 and LUX we are beginning to better understand the interactions of impurities between the liquid and gas phase which will prove valuable for designing of next generation ton-scale liquid xenon experiments.

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