

Cryogenic Ion Capture Probe R&D at SLAC

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Abstract

During the early R&D phase for EXO-200 starting in 2001, the SLAC group designed and built a liquid xenon(LXe) ‘probe test cell’. This setup was used at first for thorium ion mobility measurements described in [1]. In addition, the setup was used for barium tagging R&D, testing several barium ion capture ‘probe’ designs, including in May 2003 a novel cryogenic probe built from a modified surgical instrument. As this approach has been recently adopted for R&D work by the Colorado State (CSU) tagging program, it seems like a good time to review the origins of the method. The history and the results from the work at SLAC will be described.

1 Introduction

Once it was determined that a LXe TPC would be the default design during the earliest days of EXO R&D work circa 2000, the barium ion tagging scheme of choice became electrostatic ion capture in the liquid, followed by high-specificity ion identification. The Stanford group launched a successful barium identification program [2], while the SLAC group proceeded with R&D for electrostatic capture. To this end, a small LXe test cell was designed and built onto the already existing xenon purification and purity monitor (XPM) system.

2 The Probe Test Cell

In order to test the properties of ions in LXe, and the efficacy of various electrostatic capture schemes, the ions had to be easily identifiable. As our system would not include elaborate (and at the time untested) fluorescence tagging, this is most convenient if the ions are alpha decaying isotopes. In addition, the ions should be as similar to barium ions as is reasonably possible, and mainly this meant that the species should have a similar ionization potential. At the suggestion of Giorgio Gratta, a uranium 230 source was chosen - a source which would recoil thorium 226 ions into the liquid, and that provides a distinctive four-alpha-peak signature. The thorium first ionization energy is 6.1 eV compared to 5.2 eV for barium. The source was produced from a thorium 232 target at the UC Davis cyclotron (see <http://exo-eelog.stanford.edu/Collaboration/4>), and the nuclear chemistry required for ^{230}U source preparation (plated on to a 1 cm platinum disk) was performed at LLNL by Kevin Roberts in the summer of 2002.

The Probe Test Cell needed to satisfy a number of basic requirements :

1. A small LXe cell with view ports, and a submerged cathode and anode (horizontally arranged).
2. A source of alpha decaying ions at the anode
3. A vertically movable HV probe that could moved between three positions;
 - Immersed in the LXE,

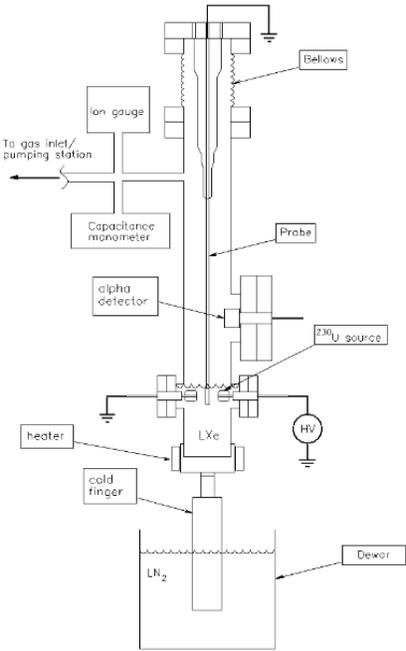


Figure 1: Schematic illustration of the probe cell. Not shown in the diagram are the two view ports located at the same height as the uranium source, 90 degrees over in azimuth, nor the precision stage that is used to vertically position the bellows/probe assembly.

- Above the L/g interface adjacent to alpha counters,
 - Above the alpha counters.
4. A set of alpha counters in the gas phase at position (2)

Additional details regarding the test cell, simple electrostatic capture probe designs and the radioactive source used are available in [1],[3]. In terse summary, the arrangement outlined above allowed for the electrostatic capture, identification, and in principle the ‘release’ of the thorium ion daughters recoiled into the LXe. At the lowest probe position, the tip is located between the electrodes and Th ions in the LXe are electrostatically captured while the probe is set to a negative high voltage (HV) potential (typically -1 kV). When the probe is raised to the ‘counting’ position, the alpha decays of the captured ions are observed. The uppermost position can be used to release the ions while the probe HV potential is reversed to a positive value (eg. +1 kV). Early tests with a simple electrostatic probe for which the SS probe tip could be set to negative potential for the collection of positive ions confirmed the expectation that ions captured on a conducting surface could not be released by a simple field reversal - not surprising given the magnitude of image-charge electric fields.

3 Electrostatic Capture Probes

3.1 Early Probe Designs

In early tests, an ion capture probe including a metallic tip set to negative (-1 kV) potential was used to collect thorium ion in the LXe. As expected, once the metal ions were attached to the tip,

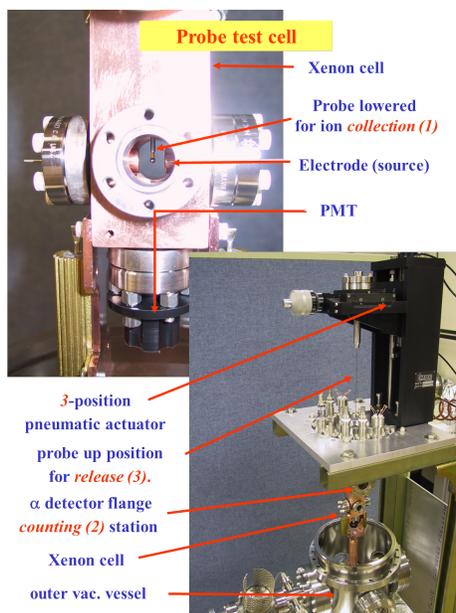


Figure 2: The electrostatic probe test cell (with an early prototype probe installed).

they could not be removed by reversing the electric field - the binding fields on the metal surface were estimated to be in the MV/cm range. This was verified by raising the probe after ion collection in LXe to the ‘count’ (2) position after having run the tip to reverse bias (+ 1 kV) and observing the unattenuated alpha decay signal. It was expected that a version of the probe with a thin dielectric coating on the tip would have the same problem due to the proximity of the image charge in the conductor, and this too was verified.

Figure 2 shows a photo of the probe test cell with an early electrostatic probe installed. The upper image details the source and electrodes with the probe in the lowest position for ion collection, the lower image shows the outer cryostat vacuum vessel retracted below the cell and the precision pneumatic stage used for probe positioning at the three probe positions above. The alpha decays of captured radioactive ^{226}Th ions provided an unambiguously distinctive signature as can be seen in Figure 3.

3.2 Cryoprobe Designs

We then proposed that a way to avoid irreversible ion capture would be to instead have the tip coated with a thin layer of xenon ice. Ion removal would be accomplished by sublimation of this ice in xenon gas at a pressure below which there is no liquid phase - for xenon 614 torr. While alternatives were considered (Peltier effect solid state coolers, for example, are not efficient at LXe temperature), the most straightforward tip-cooling technique is Joule-Thompson (J-T) constant enthalpy expansion of the appropriate gas. The inversion temperature, below which J-T expansion results in gas cooling, is high for most gases (798 K for argon, 640 K for nitrogen), and low for some (35 K for Helium).

3.3 Modeling Joule Thompson Cooling

The expected performance of the cryo-probes can be estimated from the basic principles of the J-T process. The calculations made in 2003 are summarized here.

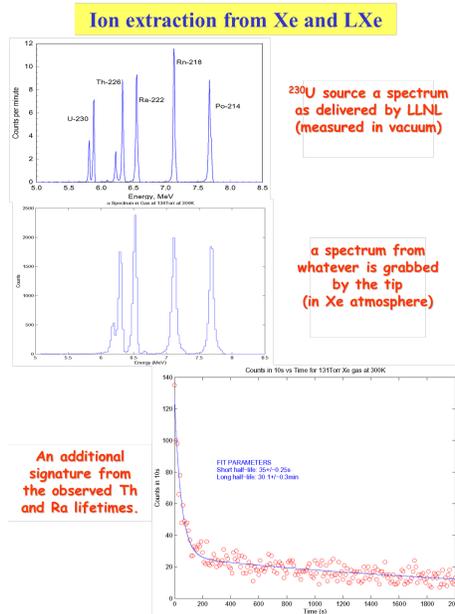


Figure 3: The alpha detector response in vacuum provided by the ^{230}U source supplier, and the observed spectrum from captured thorium daughters extracted from the LXe as seen by the silicon surface-barrier alpha detectors in the xenon gas phase above the liquid. Also shown is the observed half life corresponding to the thorium isotope.

The Joule Thompson coefficient for constant enthalpy cooling is defined as

$$\mu = \left(\frac{dT}{dP} \right)_H, \quad (1)$$

where the subscript H indicates constant enthalpy ($H = U + PV$). For an ideal gas, this coefficient is identically zero. Non-ideal behavior including the effects of intermolecular forces can be parameterized by the so-called Virial Expansion in powers of the number density of the gas, the first two terms of which appear as

$$P = kT(n + B_2(T)n^2 + \dots) \quad (2)$$

where $B_2(T)$ is the second virial coefficient. The virial coefficients are approximately calculable when a molecular interaction potential is added to the Hamiltonian and is included in the calculation of the partition function. The second virial coefficient shown here can be connected to the modifying terms in the Van der Waals extension of the ideal gas law. One parameterization, given in Figure 4 was taken from textbook data [4] for argon, and models intermolecular forces. The temperature dependence of the J-T coefficient can then be obtained from

$$\mu = \frac{N_0}{c_P} \left(T \frac{dB_2(T)}{dT} - B_2(T) \right). \quad (3)$$

The result for argon is shown in Figure 4. A custom made J-T cooled probe was built and tested using both high pressure argon and helium (see Figure 5). The data is shown in Figures 6,7, with the expected behavior seen (cooling with argon, warming with helium), however the cooling power was too low to be useful with this probe.

Remarkably, a commercial product was found that, with only minor modifications, was a very good fit to our needs. Produced at the time by two different companies Endocare Inc. and Galil

Joule-Thompson (constant enthalpy) cooling

$$\mu = \left(\frac{dT}{dP} \right)_H$$
 The Joule-Thompson coefficient for real gases changes sign at an inversion temp. T_{inv} (eg. 640 °K for N_2 , 35 °K for He)

$$\mu = \frac{N_0}{c_p} \left(T \frac{dB_2(T)}{dT} - B_2(T) \right)$$
 The J-T coefficient can be calculated from a modified ideal gas law by using the *Virial Expansion*. The B_2 coefficient depends on the intermolecular potential.

$$P = kT(n + B_2(T)n^2 + \dots)$$

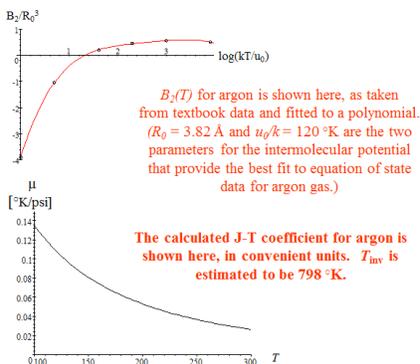


Figure 4: The second virial coefficient for argon that was used, and the resulting temperature dependent J-T coefficient for argon.

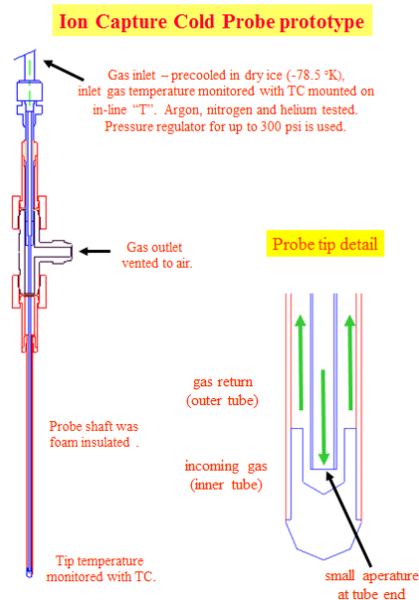
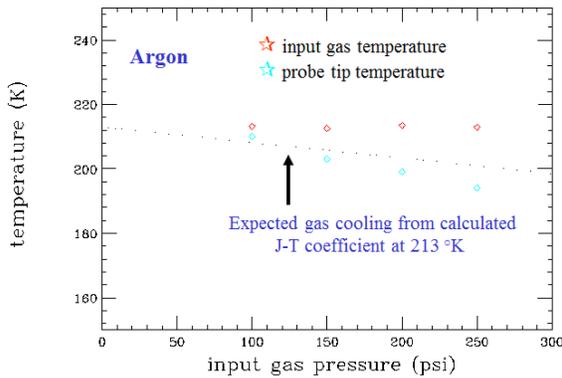
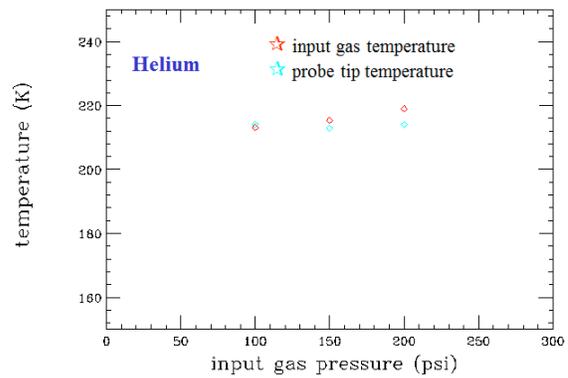


Figure 5: Illustration of homemade J-T cryoprobe.



The observed temperature drops were 3,10,14 and 19 degrees for the 100,150,200 and 250 psi input pressures. (12 degrees was the calculated J-T temperature drop for a 250 psi ΔP .)

Figure 6: Data from the homemade cryoprobe for argon gas.



The data certainly looks different than the argon data – The tip temperature doesn’t change at all, while the input temperature rises with pressure. Perhaps J-T heating is occurring as the He gas leaves the regulator.

Figure 7: Data from the homemade cryoprobe for helium gas.

Medical, cryosurgical instruments based on the J-T effect are available in small diameter ‘probe’ format. We contacted a urologist at UCSF in San Francisco, Dr. Katsuto Shinohara, who used these devices in his practice. PCR visited his office at the Comprehensive Cancer Center in San Francisco and Dr. Shinohara described and demonstrated the cryoprobes and kindly provided several Endocare and one Galil instruments (designated for disposal) free of charge. The Endocare probes came in both 2.4 mm and 3.4 mm diameter models, and were conveniently equipped with an internal thermocouple that measured the temperature reached at the probe tip (coldest location).

K. Skarpaas was able to design, and with R. Conley’s help, execute, modifications to the medical instruments from Endocare for implementation in our test setup. This included an interface with our high pressure argon system, provision for HV attachment (for electrostatic ion collection), and the addition of a vacuum jacket to localize the cooling to the probe tip area. Delicate construction of thin-walled concentric tubing and precision welding was required. Figure 8 shows an x-ray image (taken using a dental x-ray machine - we received assistance from the medical community more than once) of one such customized cryoprobe.

The 2.4 mm customized Endocare cryoprobe was studied in our setup. A big advantage of the customized cryoprobes, compared to our homemade prototype, is that much better cooling performance is observed. This is due to the very efficient miniature heat exchanger engineered into these probes, that uses the J-T cooled outgoing argon to cool the incoming gas. A semi-empirical prediction of cooling performance, using the calculated temperature-dependent J-T coefficient shown in Figure 4, was compared to data (argon gas was used). The effectiveness of the cryoprobe internal heat exchanger was not known, but the assumption that seemed to work best took the cooling (ΔT) of the incoming gas due to the miniature heat exchanger to be linear in the pressure differential (ΔP), where different choices for the lower pressure ΔT can be tried and extrapolated to higher pressure. Due to the gas flow rate also being proportional to ΔP , this leads to a cooling power quadratic in ΔP - this procedure apparently was matched to the roughly quadratic behavior of the J-T coefficient with input temperature. Reasonable agreement was seen as illustrated in Figure 9. Note that in the medical application, input gas pressures of up to 3,500 psi are used due to the heat bath environment of the probe tip during surgical procedures. Our maximum pressure of 800 psi achieved a tip temperature of 120K with the probe tip in vacuum. (It should be noted that the type T thermocouple deployed within the probe is not calibrated and is expected to read high with respect to the true temperature by a few degrees at the lowest readings.)

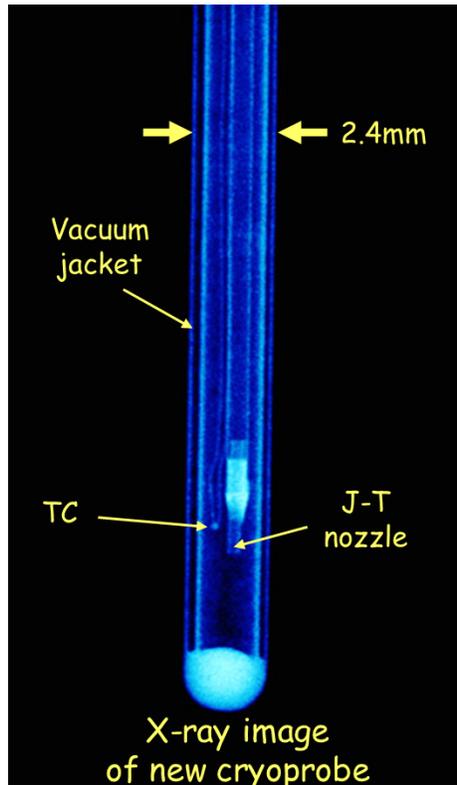


Figure 8: An x-ray of a 2.4 mm customized Endocare probe. The added vacuum jacket is visible, as are the factory provided J-T nozzle and miniature heat exchanger, and the internal TC.

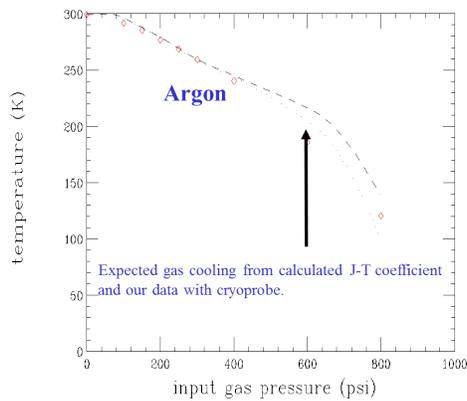


Figure 9: The temperature dependent J-T coefficient for argon derived earlier was used to estimate the cooling performance of the 2.4 mm customized cryoprobe. The two different semi-empirical curves correspond to different choices for the effectiveness of the internal heat exchanger at low input pressure. The red points are the data.

Ion capture and release was studied with this instrument and in more than one modified form. While a number of effects were observed and are described in more detail in [3], the most important observation made was that by capturing Th ions onto a xenon ice surface, it was possible to prevent irreversible adsorption of these ions onto the probe's conducting surface. Graceful release of the ions from the xenon ice was not demonstrated in these experiments due to the liquifaction of the ice when the probe tip is allowed to warm in a high pressure xenon gas environment. It was our hypothesis at the time that to do so would have required a method for reducing the xenon gas pressure to the 614 torr value below which sublimation occurs - and the setup would have required reconfiguration with a gate valve and an isolated volume for pumpdown. Barium tagging schemes that do not require Ba ion release from the xenon ice avoid this issue, and this is the approach presently under study by W. Fairbank, Jr. and the CSU group [5].

4 Conclusions

We have presented an outline of the original R&D work done at SLAC towards a scheme for barium ion collection and release in LXe, culminating in the invention and testing of a cryogenic probe based on a modified medical instrument. The utility of ion capture in xenon ice was demonstrated at the time. This work was completed over ten years ago, but the basic concept has more recently been revived in the activities of the EXO CSU group. It is possible that alternate schemes for cryoprobe cooling will be needed (directly LHe cooling, for example) in order to achieve lower xenon ice temperatures, but we speculate here that two-stage J-T schemes (argon, and hydrogen, with an inversion temperature of about 200K) or hybrid schemes (LHe cooling/J-T hydrogen) might be usable.

5 Acknowledgements

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