Accumulation and recovery of large quantities of hyperpolarized ^{129}Xe from high flow mixtures with minimum losses

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Introduction

Hyperpolarized gases offer a new high performance diagnostic contrast agent in MRI[1]. ^{3}He has been studied most extensively due to the highly refined polarization technology[2]. In contrast, technology improvements for producing, isolating, and preserving ^{129}Xe polarization are still ongoing. We developed a large-scale high-performance xenon polarizer[3] that produces as much as 1.2 liters/hr of xenon at 45% polarization. The polarized xenon is a small fraction of the high-flow low-pressure gas mixture flowing at 1.5 liters/minute with xenon flows between 0.005 and 0.05 liters/minute. A liquid nitrogen trap is used to separate xenon from helium and nitrogen buffer and quenching gases. While xenon in the frozen state can have long relaxation times[4], ~2-3 hours at 77 K, relaxation rate depends strongly on temperature and on external magnetic field[5] for temperatures above 120 K. A major issue in recovering the xenon polarization from its frozen state is the time spent near the phase transition where relaxation times can be of the order of seconds. Although recovery ratios of over 90% for small quantities have been reported[6],losses as high as 30-50% are routinely observed for large quantities.

The most common apparatus for trapping xenon from a gas mixture is the coaxial double-tube condenser shown in Fig.1. The initial gas mixture comes through the inner glass tube, starts to cool down, and freezes in a thick layer on the outer tube walls. The inner tube can be warmed to prevent freezing there (not shown). Hypotheses exist for loss mechanisms during both accumulation and thawing. During accumulation, warm gas mixture passing over already frozen xenon could maintain the top layer of frozen xenon at too warm a temperature, particularly for thick accumulations. More recent studies[7] found that thawing dominated, at least in their system. They observed that when xenon thawed and passed through liquid phase, it could just drop to the bottom of the cell and spend too long in the liquid phase. Starting from these two hypotheses, we designed and built a new xenon freeze-out system which could eliminate the fast relaxation mechanisms.

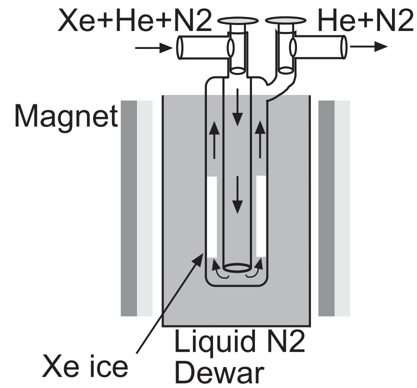

Fig.1: Straight-tube coaxial configuration of a freeze-out: xenon accumulates on the cold bottom and walls of the outer tube.

Methods

The schematic of the implemented freeze-out system with the UNH high-flow low-pressure polarizer is shown in Fig.2. With five turns of 1 cm tubing in a 15 cm height, our "spiral" freeze-out offers a large surface for xenon trapping. For the liquid nitrogen dewar we used an "U"-shaped homebuilt aluminum foam covered container. The magnet box was also built in-house using NdFeB rare-earth permanent magnets, which generate a field of 0.31 T. Iron field clamps were added to shim the field homogeneity inside the gap and to reduce the field gradients for the incoming polarized xenon on top of the box.

The working principle of our freeze-out is quite simple. Gas mixture containing hyperpolarized ^{129}Xe comes from the polarizer through a transfer tube, where ^{129}Xe polarization can be measured using an NMR coil setup. It enters the freeze-out through a stopcock and flows downwards through the spiral. We have the liquid nitrogen dewar fully filled and positioned such initially we cover just the bottom spiral of the freeze-out. Depending on how much xenon we want trapped and the accumulation time, we can gradually or periodically raise the LN2 level by raising the dewar. Xenon forms a thin layer of ice distributed over the freeze-out wall surface. At the end, the xenon is uniformly spread inside the spiral in a layer proportional with the quantity of xenon frozen. Thawing of this thin uniform layer proceeds quickly when immersed in warm water.

Results and Outlook

We measured recovery ratios of the polarization as a function of production rate for 10 minutes accumulation time, leading to different accumulated quantities. The initial polarization was measured during production of polarized xenon and the final polarization was measured in the same location, such minimizing uncertainties for recovery ratios to ~2%. The results of our study are presented in the attached Table.

Xenon Flow (liters/hour)	Frozen Volume (ml)	Initial Polarization (%)	Thawed Polarization (%)	Recovering Ratio (%)
0.6	100	47.4	47.3	99.8
3.0	500	24.5	24.4	99.6
4.5	750	19.1	17.6	92.0
6.0	1000	16.4	14.5	88.5

We demonstrated full recovery of ~100% for xenon polarization after freeze-thaw cycles for quantities up to 500 cc. Once the quantities were increased the layer of xenon ice starts to grow over the limit of keeping temperature below 120 K and the relaxation mechanism moves into a new regime, so we observed losses in the thawed polarized xenon. We are partnering with Xemed LLC to scale up this technology by a factor of ten, and automate the motion of the dewar for improved uniformity.

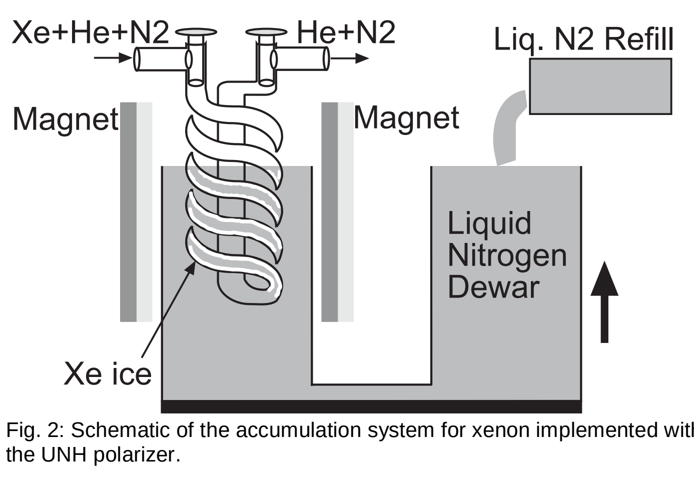

Fig. 2: Schematic of the accumulation system for xenon implemented with the UNH polarizer.

Acknowledgements

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References:

1. B.M. Goodson, *J. Mag. Res.* 155, 157-216 (2002)
2. T.G. Walker, W. Happer, *Rev. Mod. Phys.* 69, 629-642 (1997); J. Becker et al., *Nucl. Inst. Meth. Phys. Res. A* 402, 327-336 (1998)
3. I.C. Ruset et. al., *Large Production of Highly Polarized ^{129}Xe and Delivery*, the 2004 International Workshop on Pulmonary Function Imaging at Penn, Philadelphia, PA, Nov. 12-14
4. G.D. Cates et al., *Phys Rev Lett* **65**, 2591 (1990)
5. N.N. Kuzma et al., *Phys Rev Lett* **88**, 1476021-4 (2002)
6. U. Ruth et al. *Appl. Phys. B* **68**, 93-97 (1999)
7. B. Patton et al, 45th ENC, Asilomar, CA, p174 (2004)