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Chromatographic separation of radioactive noble gases from xenon

D.S. Akerib[a,b,c], H.M. Araújo[d], X. Bai[e], A.J. Bailey[d], J. Balajthy[f], P. Beltrame[g], E.P. Bernard[h], A. Bernstein[i], T.P. Biesiadzinski[a,b,c], E.M. Boulton[h], R. Bramante[a,b,c], S.B. Cahn[h], M.C. Carmona-Benitez[j], C. Chan[k], A.A. Chiller[l], C. Chiller[l], T. Coffey[a], A. Currie[d], J.E. Cutter[m], T.J.R. Davison[g], A. Dobi[n], J.E.Y. Dobson[o], E. Druszkiewicz[p], B.N. Edwards[h], C.H. Faham[n], S. Fiorucci[k,n], R.J. Gaitskell[k], V.M. Gehman[n], C. Ghag[o], K.R. Gibson[a], M.G.D. Gilchriese[n], C.R. Hall[f], M. Hanhardt[e,q], S.J. Haselschwardt[j], S.A. Hertel[r,h], D.P. Hogan[r], M. Horn[r,h], D.Q. Huang[k], C.M. Ignarra[b,c], M. Ihm[r], R.G. Jacobsen[r], W. Ji[a,b,c], K. Kamdin[r], K. Kazkaz[i], D. Khaitan[p], R. Knoche[f], N.A. Larsen[h], C. Lee[a,b,c,*], B.G. Lenardo[m,i], K.T. Lesko[n], A. Lindote[s], M.I. Lopes[s], A. Manalaysay[m], R.L. Mannino[t], M.F. Marzioni[g], D.N. McKinsey[r,h], D.-M. Mei[l], J. Mock[m], M. Moongweluwan[p], J.A. Morad[m], A.St.J. Murphy[g], C. Nehrkorn[j], H.N. Nelson[j], F. Neves[s], K. O'Sullivan[r,n,h], K.C. Oliver-Mallory[r], K.J. Palladino[v,b,c], E.K. Pease[h], K. Pech[a], P. Phelps[a], L. Reichhart[o], C. Rhyne[k], S. Shaw[o], T.A. Shutt[a,b,c], C. Silva[s], V.N. Solovov[s], P. Sorensen[n], S. Stephenson[m], T.J. Sumner[d], M. Szydagis[u], D.J. Taylor[q], W. Taylor[k], B.P. Tennyson[h], P.A. Terman[t], D.R. Tiedt[e], W.H. To[a,b,c], M. Tripathi[m], L. Tvrznikova[h], S. Uvarov[m], J.R. Verbus[k], R.C. Webb[t], J.T. White[t], T.J. Whitis[a,b,c], M.S. Witherell[j], F.L.H. Wolfs[p], K. Yazdani[d], S.K. Young[u], C. Zhang[l]

[a] *Case Western Reserve University, Dept. of Physics, 10900 Euclid Ave, Cleveland OH 44106, USA*
[b] *SLAC National Accelerator Laboratory, 2575 Sand Hill Road, Menlo Park, CA 94025, USA*
[c] *Kavli Institute for Particle Astrophysics and Cosmology, Stanford University, 452 Lomita Mall, Stanford, CA 94309, USA*
[d] *Imperial College London, High Energy Physics, Blackett Laboratory, London SW7 2BZ, UK*
[e] *South Dakota School of Mines and Technology, 501 East St Joseph St., Rapid City SD 57701, USA*
[f] *University of Maryland, Dept. of Physics, College Park MD 20742, USA*
[g] *SUPA, School of Physics and Astronomy, University of Edinburgh, Edinburgh EH9 3FD, United Kingdom*
[h] *Yale University, Dept. of Physics, 217 Prospect St., New Haven CT 06511, USA*
[i] *Lawrence Livermore National Laboratory, 7000 East Ave., Livermore CA 94550, USA*
[j] *University of California Santa Barbara, Dept. of Physics, Santa Barbara, CA, USA*
[k] *Brown University, Dept. of Physics, 182 Hope St., Providence RI 02912, USA*
[l] *University of South Dakota, Dept. of Physics, 414E Clark St., Vermillion SD 57069, USA*
[m] *University of California Davis, Dept. of Physics, One Shields Ave., Davis CA 95616, USA*
[n] *Lawrence Berkeley National Laboratory, 1 Cyclotron Rd., Berkeley CA 94720, USA*
[o] *Department of Physics and Astronomy, University College London, Gower Street, London WC1E 6BT, UK*
[p] *University of Rochester, Dept. of Physics and Astronomy, Rochester NY 14627, USA*
[q] *South Dakota Science and Technology Authority, Sanford Underground Research Facility, Lead, SD 57754, USA*
[r] *University of California Berkeley, Department of Physics, Berkeley CA 94720, USA*
[s] *LIP-Coimbra, Department of Physics, University of Coimbra, Rua Larga, 3004-516 Coimbra, Portugal*
[t] *Texas A & M University, Dept. of Physics, College Station TX 77843, USA*
[u] *University at Albany, State University of New York, Department of Physics, 1400 Washington Ave., Albany, NY 12222, USA*
[v] *University of Wisconsin-Madison, Department of Physics, 1150 University Ave., Madison, WI 53706, USA*

* Corresponding author at: Center for Underground Physics, Institute for Basic science (IBS), Daejeon 305-811, Republic of Korea
E-mail address: cxl370@case.edu (C. Lee).

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ABSTRACT

The Large Underground Xenon (LUX) experiment operates at the Sanford Underground Research Facility to detect nuclear recoils from the hypothetical Weakly Interacting Massive Particles (WIMPs) on a liquid xenon target. Liquid xenon typically contains trace amounts of the noble radioactive isotopes ^{85}Kr and ^{39}Ar that are not removed by the *in situ* gas purification system. The decays of these isotopes at concentrations typical of research-grade xenon would be a dominant background for a WIMP search experiment. To remove these impurities from the liquid xenon, a chromatographic separation system based on adsorption on activated charcoal was built. 400 kg of xenon was processed, reducing the average concentration of krypton from 130 ppb to 3.5 ppt as measured by a cold-trap assisted mass spectroscopy system. A 50 kg batch spiked to 0.001 g/g of krypton was processed twice and reduced to an upper limit of 0.2 ppt.

1. Introduction

Liquid xenon is an excellent target for the direct detection of WIMP dark matter [1], particularly when instrumented in a time projection chamber (TPC) as in the LUX detector [2]. Xenon's high proton number allows a very short penetration depth of external gamma and beta radiation [3,4], and the event-by-event position measurement of the TPC allows these backgrounds to be highly suppressed in the inner volume of the detector. Because it has no long-lived radioactive isotopes [5], xenon is intrinsically quiet. The average single-scatter rate in the energy window of 0.9–5.3 keV$_{ee}$[1] inside the 118 kg fiducial mass in LUX is measured to be below 10^{-3} events per kg/day/keV (differential rate unit, DRU$_{ee}$) [6]. This rate is dominated by the gamma rays from radioactive impurities in the 122 Hamamatsu R8778 photomultiplier tubes (PMTs) [7].

Xenon, being distilled from the atmosphere, contains noble radioactive impurities such as ^{85}Kr and ^{39}Ar with half-lives of 10.756 yrs and 269 yrs, respectively. [8,9]. Their characteristics are summarized in Table 1.

^{85}Kr is generated by anthropogenic fission, and released into the atmosphere primarily during nuclear fuel reprocessing [10]. It contributes about 1 Bq/m^3 of the radioactivity from atmosphere [11], from which one can deduce that about 10 parts-per-trillion (ppt, 10^{-12}) (g/g)[2] of atmospheric Kr is ^{85}Kr. A measurement based on low-level counting reported 4–22.5 ppt [12]. Research-grade xenon contains about 10^{-7} natural krypton by mass. One analysis of boil-off gas of a distillation tower revealed that 6 ± 2 ppt of the krypton impurity is ^{85}Kr in their sample [13]. At these concentrations, the decay of ^{85}Kr yields a rate of about 6 DRU$_{ee}$, which overwhelms the potential dark matter signal. To be comparable to the background rate due to the PMTs, the xenon in LUX must contain less than 20 ppt krypton. The goal for LUX to reduce the ^{85}Kr concentration to below 4 ppt was met using the method described in this paper.

^{39}Ar is mainly generated by the $^{40}\mathrm{Ar} + \mathrm{n} \rightarrow {}^{39}\mathrm{Ar} + 2\mathrm{n}$ process in the atmosphere due to cosmic rays, and constitutes about 1 ppq of argon in the atmosphere [9]. The isotope emits a beta particle with an endpoint of 0.565 MeV. The LUX research-grade xenon originally contained about 1 ppb of argon. However, a portion of xenon was retrieved from other experiments, and contained an unknown amount of argon. For its background rate to be comparable to that from the photomultiplier tubes in the fiducial volume, the argon concentration must be reduced below ppb.

Cryogenic distillation has been used to separate these light radioactive noble impurities from xenon. The XMASS detector is a single-phase dark matter detector containing 800 kg of liquid xenon. The XMASS collaboration developed a cryogenic distillation column to reduce the krypton level in xenon. Their distillation column lowered the krypton concentration by a factor of 1,000 to 1.9 ppt of krypton [14]. A similar system was adopted by the XENON100 collaboration [15], and dropped their krypton concentration below 1 ppt [16].

2. Adsorption-based chromatography

Adsorption-based chromatography is widely used for gas separation in industrial and chemical applications. A common industrial application is the separation of nitrogen from air, known as "pressure swing adsorption." Among the scientific applications, the Borexino [17] and NEMO-2 [18] collaborations developed a charcoal adsorption system to remove atmospheric radon from underground laboratory air, while a similar system removed krypton from xenon for the XENON-10 experiment [19]. The latter system processed 20 kg of xenon in 2 months, with the final krypton concentration below 3 ppt. In this section, we present a mathematical treatment of adsorption-based gas chromatography central to our application, following the approach presented in [17]. More general reviews are available in the literature [20].

Adsorption refers to adhesion of atoms or molecules on a surface. The typical binding energy for adsorption is smaller than that of covalent bonds, and the process is reversible:

$$R + X \rightleftharpoons RX. \tag{1}$$

Here, X is the molecule to be adsorbed, or adsorbate, on the sites provided by R, the adsorbent.

The equilibrium between the free and the adsorbed states of Eq. (1) depends principally on a few parameters: the adsorbate, the adsorbent, the ambient temperature, and the concentration of adsorbate. A simple parametrization for mono-layer adsorption was proposed by Langmuir [21]. The fraction θ of the sites occupied is described as a function of the partial pressure P of the adsorbent:

$$\theta = \frac{KP}{1 + KP}. \tag{2}$$

K is the equilibrium constant, the ratio of adsorption and desorption rates in Eq. (1). It has a dimension of inverse pressure because the rate of adsorption is proportional to the partial pressure of X. If P is large, θ converges to 1, indicating that all sites are occupied.

At low P ($KP \ll 1$), θ depends linearly on P:

$$S = S_0\theta = S_0KP = bP. \tag{3}$$

[1] The energy window calibrated with electronic recoils.

[2] Concentrations are quoted by the ratio of masses unless quoted otherwise. Parts per million (10^{-6}), billion (10^{-9}), trillion (10^{-12}), and quadrillion (10^{-15}) are abbreviated as ppm, ppb, ppt, and ppq.