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Distillation of liquid xenon to remove krypton

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ABSTRACT

A high performance distillation system to remove krypton from xenon was constructed, and a purity level of Kr/Xe = $\sim 3 \times 10^{-12}$ was achieved. This development is crucial in facilitating high-sensitivity low-background experiments such as the search for dark matter in the universe.

1. Introduction

Liquid xenon is one of the most attractive materials for use in detectors for astroparticle and particle physics [1–5]. As a scintillator it has a large light yield, comparable to that of NaI(Tl), and can be used for detectors with low energy thresholds and high energy resolution. Because of the high atomic number of xenon ($Z = 54$) and its high density in liquid form (~ 3 g/cm^3), it contributes to the reduction of environmental backgrounds, such as γ-rays and β-rays from uranium and thorium contamination, by self-shielding. Another big advantage of liquid xenon is that xenon does not have long-lived radioactive isotopes, and thus experiments on rare phenomena (such as dark matter searches and double beta decay experiments) may be carried out shortly after moving the xenon underground from the surface.

One drawback of liquid xenon is the possibility of contamination with radioactive ^{85}Kr. Xenon is usually produced from air. The concentration of xenon in air is $\sim 10^{-7}$ mol/mol, while the concentration of krypton is $\sim 10^{-6}$ mol/mol.

In the commercial production of xenon, krypton is removed by distillation or adsorption. But, the commercially available xenon contains 10^{-9}–10^{-6} mol/mol of krypton, because the removal

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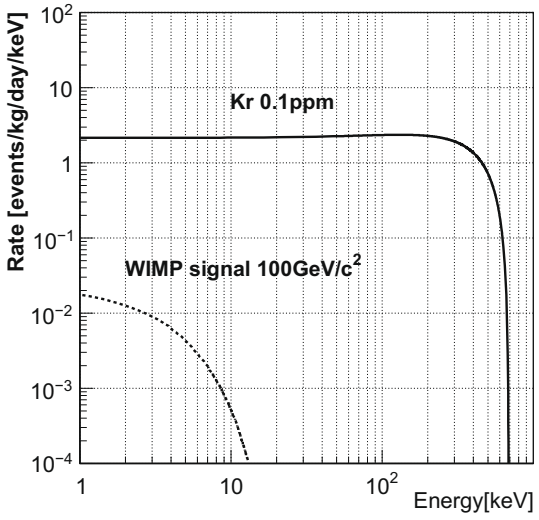

Fig. 1. Background event rate of ^{85}Kr for a Kr/Xe ratio of 10^{-7} mol/mol, compared with dark matter signal rate, assuming a cross section of 10^{-7} pb and a WIMP mass of 100 GeV/c^2. A quenching factor of 0.2 is assumed for the dark matter signal.

starts from the Kr/Xe ratio of about 10 and such purity is enough for most of the applications of xenon. ^{85}Kr is a radioactive nucleus which decays into rubidium-85 with a half-life of 10.76 years, and emits β-rays with a maximum energy of 687 keV and a 99.57% branching ratio. (The remaining 0.43% represent a β-emission with a maximum energy of 173 keV followed by a 514 keV γ-ray emission.) Large quantities of ^{85}Kr are produced artificially in nuclear fission. This constitutes the main source of ^{85}Kr in air. The concentration of ^{85}Kr in air is measured to be ∼1 Bq/m^3 [6,7], which corresponds to ^{85}Kr/Kr = $\sim 10^{-11}$. Assuming a Kr/Xe ratio of 10^{-7} mol/mol, the background event rate of ^{85}Kr is as shown in Fig. 1. (^{85}Kr/Kr of 1.2×10^{-11} is used here.) The expected dark matter event rate is also plotted, assuming a cross section of 10^{-7} pb and a WIMP mass of 100 GeV/c^2. A quenching factor of 0.2 is assumed for the dark matter signal.

Because next-generation dark matter detectors aim at a sensitivity of 10^{-9} pb for the dark matter–proton cross section, a background rate of 10^{-4} events/keV/day/kg is required. This corresponds to a Kr/Xe ratio of less than $\sim 10^{-12}$ mol/mol.

Possible methods to remove krypton from xenon are distillation and adsorption. They are commonly used industrial processes but systems which reduce Kr enough to meet our requirements did not exist. A development of adsorption-based chromatography to achieve the required Kr concentration is reported in Ref. [8].

In this paper, we describe the development of a distillation system to reduce krypton to a level of 10^{-12}. In Section 2, the design principle is discussed, while in Section 3, the setup and operation of the distillation system are described. In Section 4, we describe a technique for measuring low levels of krypton in xenon gas, and discuss the results.

2. Design principle

The boiling point of liquid krypton is 120 K at 1 atmosphere, while that of xenon is 165 K. This means that, in principle, separation of krypton and xenon can be performed by a distillation method. For the basic design of the distillation system, we followed the McCabe–Thiele (M–T) method [9].

It is the standard method of designing a distillation system. At first, the concept of the M–T method is reviewed, and then the application of the method for our purpose is described.

The principle of the M–T method is shown in Fig. 2. The main element in the distillation system is a tower in which gas–liquid equilibrium is maintained. A vessel called a "reboiler", at the bottom of the tower, boils the liquid using a heater. In order to maintain a constant temperature profile in the tower, a condenser is placed at its top. The supplied xenon gas is cooled down to near boiling point, and then supplied to the feed point in the tower at a flow rate F, as shown in the figure. The processed xenon, which contains a lower concentration of krypton than the original xenon, is obtained from the reboiler (flow rate W), and xenon with a higher krypton concentration is obtained at the top of the tower (flow rate D). In the following, the Kr concentration of xenon at these points is expressed as x_F, x_W and x_D, respectively. The heating power of the reboiler and the cooling power of the condenser at the top of the tower control the flow of xenon in the tower (flow rate L). The reflux ratio, represented by $R = L/D$, indicates the amount of xenon returned to the tower compared with the amount extracted with a high concentration of krypton.

In the M–T method, the distillation tower is assumed to be a connected series of theoretical cells, with the gas–liquid equilibrium

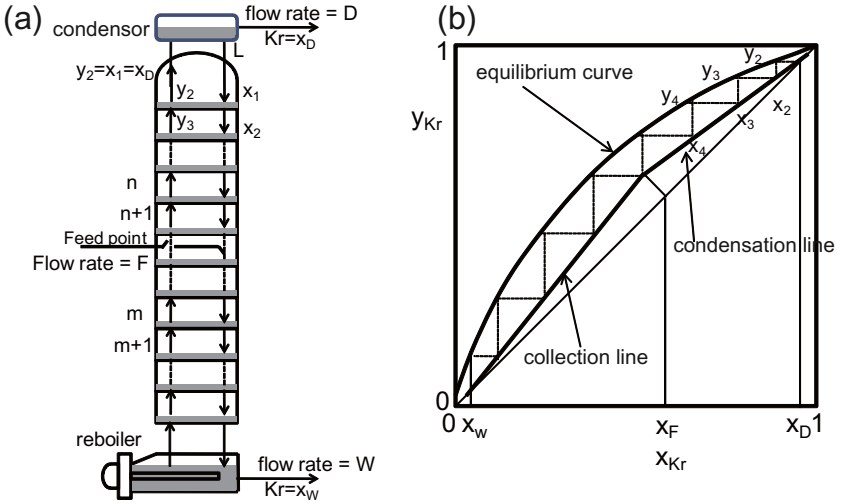

Fig. 2. (a) Illustration of the McCabe–Thiele (M–T) method. The various elements of the figure are explained in the text. (b) Calculation of the number of theoretical cells using the M–T method. The horizontal and vertical axes represent the krypton concentration in the liquid and gas phases, respectively. The thick solid curve is the equilibrium curve, and the thick solid lines are the condensation line and the collection line (details explained in the text).