A cryogenic distillation column for the XENON1T experiment

S Rosendahl1, E Brown1, I Cristescu2, A Fieguth1, C Huhmann1, M Murra1 and C Weinheimer1

1 Institut für Kernphysik, Wilhelm-Klemm Strasse 09, 48145 Münster, Germany
2 Karlsruher Institut für Technologie, Tritium Laboratory, Hermann Von Helmholtz-Platz 1, 76344 Eggenstein-Leopoldshafen, Germany
E-mail: rosendahl@wwu.de

Abstract. The XENON collaboration aims for the direct detection of cold dark matter in form of weakly interacting massive particles (WIMPs). A dual phase time projection chamber filled with liquid xenon is used to detect the WIMP-nucleon interaction. For the next generation experiment XENON1T with an active target mass of 1 ton of xenon, a new distillation column to remove krypton out of xenon to a concentration of \(< 5 \times 10^{-13}\) (0.5 ppt) natural krypton in xenon is designed and tested at the Institut für Kernphysik, Universität Münster. The experimental setup together with two diagnostic tools is presented, as well as one stability test of a 11 hour distillation run at the designed flowrate of 3kg per hour.

Keywords: Xenon; Krypton; Cryogenic distillation; Ultra-pure noble gases; Dark matter

1. Introduction
A various number of astronomical observations implies, that the matter content of the universe is dominated by non-luminous, non-baryonic dark matter, while the baryonic matter only makes less than 20% of the matter [1]. One candidate for the dark matter, coming from particle physics, is the WIMP, a weakly interacting massive particle. A well motivated WIMP particle is described by super-symmetric enhancements of the standard model of particle physics, which is the lightest supersymmetric particle (LSP or neutralino). In different models the neutralino is supposed to be stable and allows the interaction with ordinary matter on the weak scale [2]. In the recent years different experiments tried to detect the interaction between cosmic WIMPs and baryonic matter, looking for energy deposition in earth bound detectors.

The XENON collaboration, searching for detecting WIMP-nucleon interactions, uses a dual-phase time projection chamber filled with xenon, while the xenon itself is the target-mass for the interaction [3]. The detection principle is shown in figure 1. The interaction of an incoming particle with the xenon is producing prompt scintillation light, called S1, as well as electrons in the liquid xenon. The charges are drifted in an electric field to the gas phase. In a second, much stronger field the electrons are extracted into the gas phase and accelerated, producing a second light signal, called S2. The ratio of S1 and S2 allows to discriminate between electronic recoils coming from interactions with electrons or gammas and nuclear recoils that are produced by neutron scattering or the WIMP-nucleon interaction.

Currently the XENON collaboration is running the XENON100 experiment in the Gran Sasso Underground Laboratory, using 62 kg of xenon as detector material, while in parallel the next
Figure 1. Working principle of the XENON100 TPC. This diagram shows how the signals are generated in the XENON100 dual phase TPC. An incoming particle produces charge and light in liquid xenon (S1), while the charges are guided in an electric field to the gas phase, where they are extracted to produce a second light pulse (S2). The photons are detected using photomultiplier tubes. The ratio of the S2 signal to the S1 signal allows to distinguish between nuclear recoil (WIMP) events and electronic recoil events [3].

generation, called XENON1T with an active detector mass of 1 ton is being build up in the same underground lab. XENON100 was the leading experiment for direct dark matter search, setting the most stringent limit in 2012 for spin-independent WIMP-nucleon cross section of $2 \times 10^{-45}$ cm$^2$ at 55 GeV/c$^2$ and 90% confidence level [4]. This result has recently been confirmed by another liquid xenon experiment, LUX, which has used an increased sensitivity to set a limit of $7.6 \times 10^{-46}$ cm$^2$ at 33 GeV/c$^2$ with 90% confidence level [5].

The XENON1T experiment is designed for a sensitivity to the WIMP-nucleon cross section of $2 \times 10^{-47}$ cm$^2$, which is more than one order of magnitude better sensitivity than current experiments. To achieve this goal, the radioactive background has to be reduced compared to actual experiments. Beside investigation of the proper detector materials, it is also necessary to clean the xenon itself from intrinsic radioactive contaminations. One of them is the $^{85}$Kr isotope, which is a $\beta$-source with an endpoint energy of 687 keV and a half-life of 10.76 years. For XENON1T it has been calculated, that the concentration of natural krypton in xenon has to be reduced below 0.5 ppt ($10^{-12}$ mol/mol), where its decay does not contribute to the background.

2. Separation of krypton and xenon using cryogenic distillation

To reach the needed concentration of natural krypton in xenon, the technique of cryogenic distillation is used. Due to the different vapor pressures of krypton and xenon at a fixed temperature, e.g. -98°C, a separation can be achieved, while the xenon gets liquefied and the krypton stays in the gas phase.

To achieve a certain separation, a series of distillation stages can be implemented. The number of distillation stages can be calculated using the McCabe-Thiele method [6]. Commercially available xenon of highest purity has a concentration of 10 ppb ($10^{-9}$ mol/mol) to 1 ppm ($10^{-6}$ mol/mol) which implies, that a separation factor of $10^4$ to $10^6$ is needed to reduce the overall amount of krypton in xenon down to 0.5 ppt.

To reach that high separation factors it has been calculated that $\sim 10$ theoretical distillation stages are needed. For the technical realization it has been decided to construct a package
column and to use structured package material (company Sulzer, type EX), as shown in figure 2 (right) to achieve a huge surface, where the distillation process takes place.

The amount of package material, that is needed, can be estimated from the height equivalent value for one theoretical distillation stage (HETP value) multiplied with the number of distillation stages. For this column a package height of $\sim 3$ m has been estimated, also including a huge uncertainty in the HETP value, since it is unknown for krypton-xenon mixtures at low concentrations. The schematic view of a package type column is also shown in figure 2 (left). In the bottom, a reservoir is mounted, where the liquid xenon is collected, the so called reboiler, while on the top of the package material a condenser is mounted which supplies a certain cooling power to the system. By heating in the bottom and cooling in the top, a stream of xenon through the package is produced. On the top of the column, the krypton enriched xenon is collected. A small fraction, called offgas, is extracted, while most of the xenon is liquefied again and streaming down to the package (reflux). The purified xenon is extracted from the liquid in the reboiler. The column for XENON1T is designed for a throughput of 3 kg/h with a krypton enriched offgas flow of only 1%. The reflux ratio of 191 is quite high to ensure a good separation efficiency.

3. Technical design of the distillation column
Since there are a lot of open questions, concerning the HETP values and the McCabe-Thiele method for low concentration-separation, it has been decided to split the project in two phases. Phase 1 foresees a distillation plant with only 1 m package material inside, with the advantage of a faster stabilization, which allows to run several performance tests on a shorter time scale, to answer some of the unknown issues.

The incoming xenon is liquefied in a first condensation station and is then injected to the package. The cryo cooler (Typ Leybold CP140T) in top of the column is providing a cooling
Figure 3. Phase-1 Distillation Column. This picture shows the final setup for the 1-m distillation column as it was planned for the first performance tests of the improved distillation design for XENON1T.

power of 200 W at -98°C, to liquefy the upstreaming xenon. The purified xenon is extracted from the liquid in the reboiler and passes a heat exchanger to evaporate and to pre-cool the incoming gas. The krypton enriched offgas is extracted at the top. The Phase-1 setup can be seen in figure 3.

In order to avoid a contamination of the xenon, the system has to be extremely leak-tight and clean. Therefore, all components have been designed to fulfill ultra-high vacuum criteria. All parts have been made of stainless steel and OFHC copper. The pipings are done with electro polished pipes, using VCR connectors, CF flanges or orbital welding technique for leak-tight connections and all parts have been cleaned using ultrasonic cleaning. Before filling any xenon to the system, the setup has been pumped, using an oil-free turbo-molecular pumping station with a scroll pump and especially the package tube with the large surface has been baked out to +100°C to remove impurities. In a second cleaning step, xenon has been circulated through the system and further impurities have been flushed out and removed by the usage of a hot zirconium based getter. The whole system is equipped with various pressure and temperature sensors (PT1000 and calibrated silicon diodes) to monitor the stability of the distillation process.
4. Diagnostics
To determine the efficiency of the distillation process, different approaches are under investigation. In the first method, a system has been designed to measure trace amounts of natural krypton in xenon using a liquid nitrogen coldtrap to enhance the sensitivity of a commercial residual gas analyzer based on a quadrupole mass filter. In the second approach, xenon is doped with a radioactive $^{83m}$Kr tracer at the inlet of the column and the activity at the outputs is detected. Both approaches will be briefly presented.

4.1. Detection of krypton in xenon using a quadrupole mass filter
In order to determine the separation efficiency of the distillation plant one has to measure the concentration of krypton at the inlet of the column and at the purified outlet. Since this column works in a regime of very low concentrations of krypton in xenon (sub-ppb range), commercial residual gas analyzers are not able to detect them in the vicinity of the xenon atoms. Therefore, the sensitivity has to be increased. In addition to the distillation plant, a system has been designed, which allows to measure krypton in xenon to a concentration of 40 ppt following an idea of reference [7]. The principle is briefly explained in the following:

To enhance the sensitivity we use again the effect of vapor pressure for the different gases at cryogenic temperatures. The major amount of the xenon sample is frozen out in a liquid nitrogen coldtrap, while the traces of krypton stay in the gas phase and pass the coldtrap nearly unattached. This gas has an increased krypton to xenon ratio and is guided through a differential pumping section into the analyzing chamber, where it can be measured by a commercial rga.

The details of the system are presented in [8].

4.2. $^{83m}$Kr-Tracer method to investigate the distillation performance
Beside the rga method, a second technique is developed, which allows a continuous monitoring of the distillation process. Since the rga method only allows to take dedicated samples and analyze them on a timescale of $\sim$1 hour, it is not feasible to monitor the dynamic of the system continuously over a long time period.

Figure 4. $^{83m}$Kr decay detector. Left: Photomultiplier tube mounted with a Teflon holder to a CF-40 feed-through flange to provide high voltage and signal read-out. Right: PMT finally positioned inside a CF-40 T-piece, where the xenon is flushed through in front of the detector.

The idea is to inject trace amounts of the radioactive krypton isomere $^{83m}$Kr to the xenon at the inlet of the column and to monitor the decays at both outlets of the column. Since $^{83m}$Kr decays with a half-live of 1.8 h to $^{83}$Kr, which is stable, there is no danger of spoiling the xenon with other long-lived radioactive impurities [9]. The concentration of $^{83m}$Kr in xenon is doped to the order of $\sim 10^{-15}$, which allows to observe the distillation process on a sub-ppt scale, due
to the fact, that $^{83m}$Kr should follow the same chemical behavior concerning the distillation process as the other krypton isotopes. The decay products (conversion- and Auger electrons as well as $\gamma$-rays) are reacting with the xenon and produce scintillation light of 171 nm wavelength [10] that can be detected using special photomultiplier tubes with optimized quantum efficiency for that region. The rate on the photomultiplier tubes is proportional to the concentration of $^{83m}$Kr in the xenon.

**Figure 5. Performance test on the Phase-1 distillation plant.** These plots show the key parameters of the distillation setup during one longterm distillation test of 11 hours under the design parameters.
The technical realization of these $^{83m}$Kr-decay detectors is shown in figure 4. The performance of these detectors is presented in a dedicated paper [11].

5. Performance studies
In the following, the first performance studies will be presented. After the cleaning procedures, the column is cooled down by starting the coldhead with a set-point temperature, allowing the xenon to liquefy at the top. As consequence, the liquid droplets stream down, cooling the whole column from top to bottom. By adding more xenon, a liquid reservoir is build up in the reboiler, which is later needed to perform the distillation. The total load inside the column is $\sim 5$ kg. The regulation during the distillation process is designed to keep the pressure inside the column constant by varying the heater power using a PID regulation algorithm.

In figure 5 the key parameters for a distillation test of 11 hours are presented. The distillation process started after 20 minutes, when the input flow (FIC01) is slowly increased to the design value of 8.4 slpm (standard liter per minute). The liquid out flow at the bottom is also increased to keep the mass balance of incoming and outgoing fluxes of the system equalized. The system needs about 100 minutes to get to the equilibrium and running stable for several hours. The pressure in the system is stable at the set point for this measurement of 1.9 bar.

It has been demonstrated so far, that the Phase-1 column is thermodynamically stable under the designed flow- and pressure parameters. The investigation of the separation efficiency started recently using the rga system, and the $^{83m}$Kr tracer method.

Acknowledgments

Different aspects of theses experiments have been funded by DFG-Großgeräte, BMBF and Helmholtz Alliance for Astroparticle physics (HAP). We would also like to acknowledge Ondrej Lebeda from the Nuclear Physics Institute, Academy of Science of the Czech Republic, Prague for kindly providing the $^{83}$Rb generator for the $^{83m}$Kr tracer technique.

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