Universität
Münster

LowRad

low radon and low internal radioactivity

purification/distillation for next generation liquid xenon experiments

GDR Deep Underground Physics plenary meeting
LYON – 11.06.2025

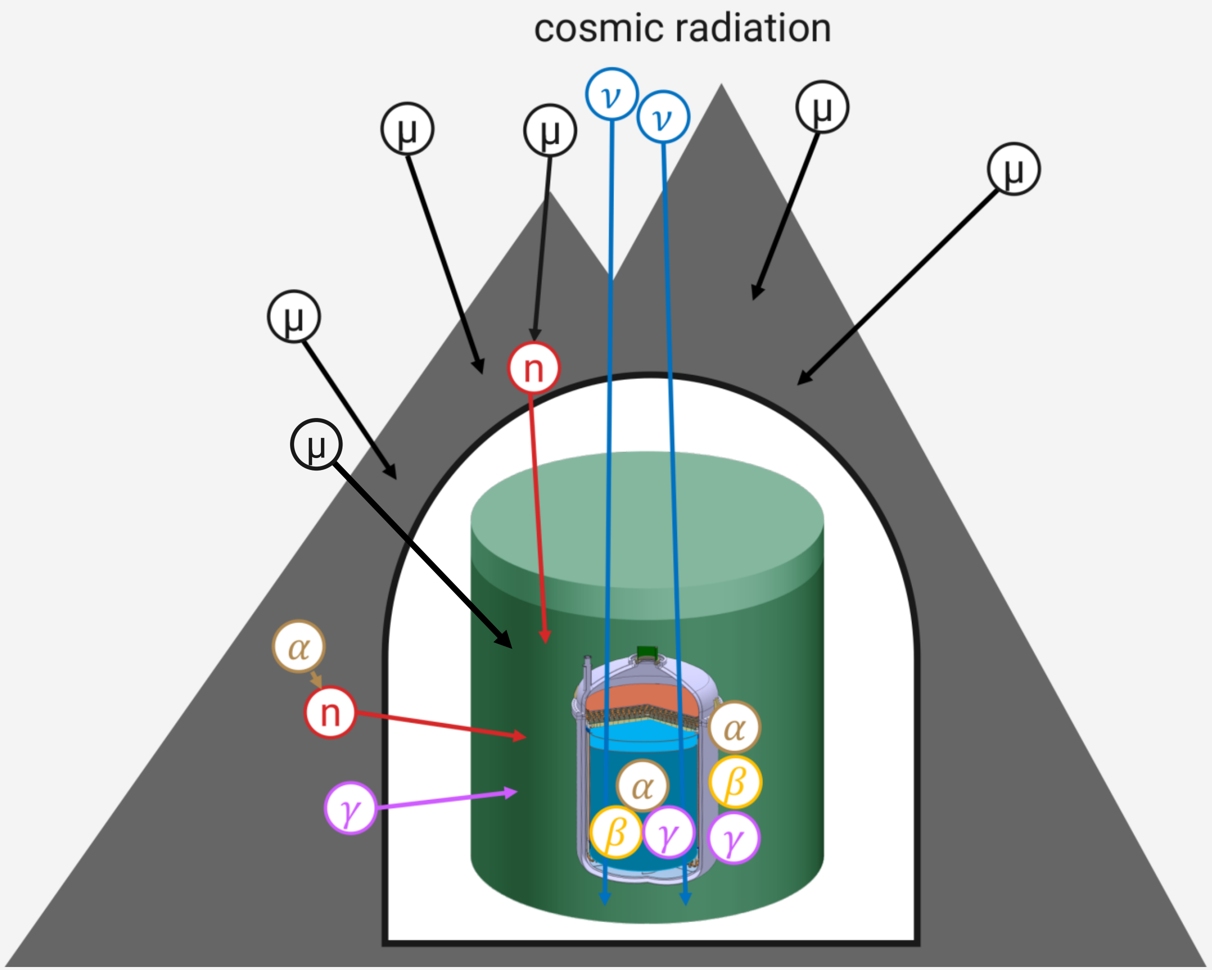
On behalf of the ERC AdG LowRad project at University of Münster:
Lutz Althüser, Robert Braun, Volker Hannen, Christian Huhmann, David Koke, Ying-Ting Lin, Philipp Schulte, Daniel Wenz, Christian Weinheimer

- Intrinsic radioactive backgrounds
- Cryogenic distillation (current status)
- Next generation: **LowRad** project

erc LowRad

living.knowledge

Limiting backgrounds in xenon DM & 0νββ detectors



Expected dark matter scattering rate:

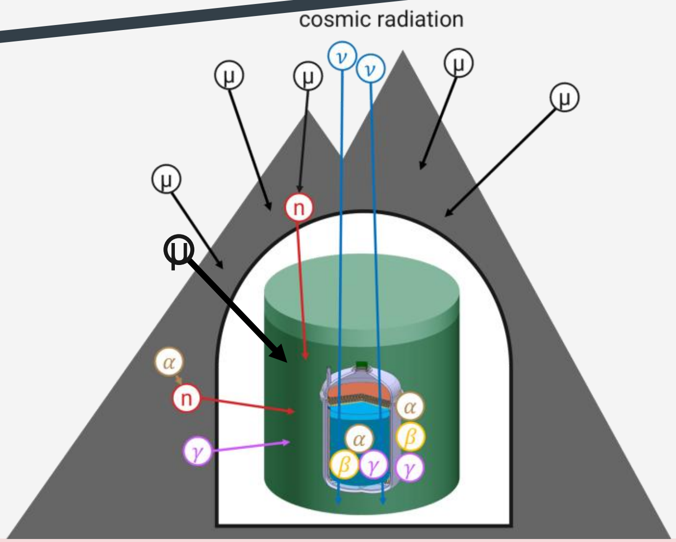
- O(0.1) event per tonne and year
- Full profit from **larger experiments only if** the experiment **remains background-free**

External background reduction:

- Underground laboratory
- Detector materials/design
- Extra (self-)shielding & muon/neutron Veto

Unavoidable backgrounds:

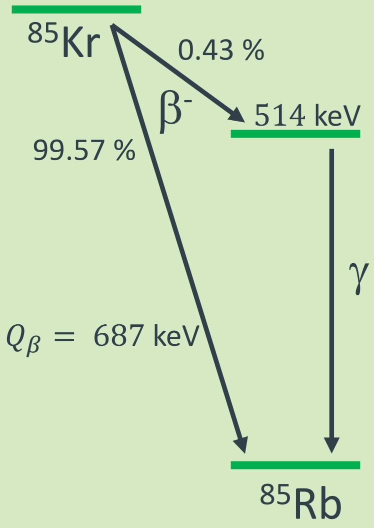
- (Solar) neutrinos
- **Intrinsic radioactive noble gas contaminants:** ^{85}Kr, ^{222}Rn and progenies, (^{37}Ar, ^{39}Ar, ^{136}Xe)
  - → ^{210}Pb plate-out (low energy searches)
  - → ^{214}Pb for DM searches
  - → ^{214}Bi for 0νββ searches



Intrinsic radioactive backgrounds

Intrinsic noble gas contaminants ^{85}Kr, ^{37}Ar and ^{222}Rn

- Leakage events from the low energy β-spectrum (ER) **contaminate** the ROI for NR DM searches
- Physics searches in the ER band only sensitive with **low background** levels



^{85}Kr

- Anthropogenic production (nuclear fission)
- Originates from Xe extraction from air
  2×10^{-11} ^{85}Kr in ^{nat}Kr
- Commercial Xe: ^{nat}Kr/Xe > 10^{-9} (ppb)

- Needs to be **removed once**
- Kr can **re-enter** via (tiny) air leaks or due to operations/maintenance

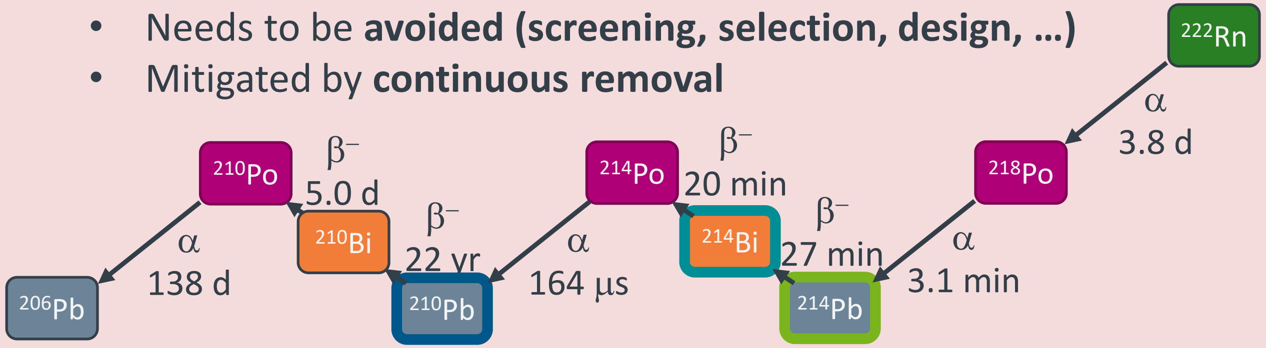
- Monitoring: Coincidence analysis, RGMS, …

Requirements for ^{nat}Kr/Xe
XENON1T: $< 0.5 \times 10^{-12}$ (0.5 ppt)
XENONnT: $< 0.2 \times 10^{-12}$ (0.2 ppt)
DARWIN/XLZD: $< 0.1 \times 10^{-12}$ (100 ppq)

^{222}Rn

- Continuous emanation from detector materials
- Background from **β-decay**, **γ-decay (0νββ)**, **plate-out**
- Main background in current and next-gen Xe experiments

- Needs to be **avoided (screening, selection, design, …)**
- Mitigated by **continuous removal**



Requirements for ^{222}Rn/Xe
XENON1T: ~10 µBq/kg (10^{-24})
XENONnT: ~1 µBq/kg (10^{-25})
DARWIN/XLZD: ~0.1 µBq/kg (10^{-26})

Further mitigation by decay tagging
Monitoring: α-analysis, Bi-Po, …

Cryogenic distillation

Difference in vapor pressure of the noble gas elements

- Brought into our field by XMASS for **Kr removal**: Astropart. Phys. 31, 290-296 (2009)
- Continued by **XENON** and **enhanced to "online" Kr & Rn removal**:
  EPJ C 77 277 (2017), EPJ C77 358 (2017), PTEP 053H01 (2022), EPJ C 82 1104 (2022), …

Relative volatility: $\boldsymbol{\alpha = P_{\mathrm{ng}}/P_{\mathrm{Xe}}}$ (@ 178 K)

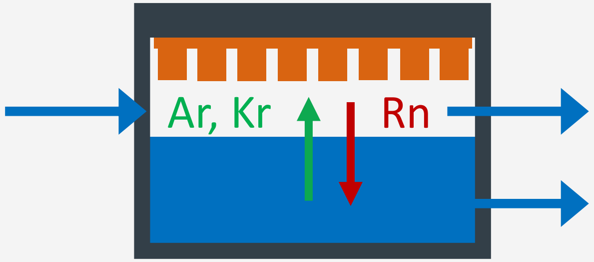
^{85}Kr

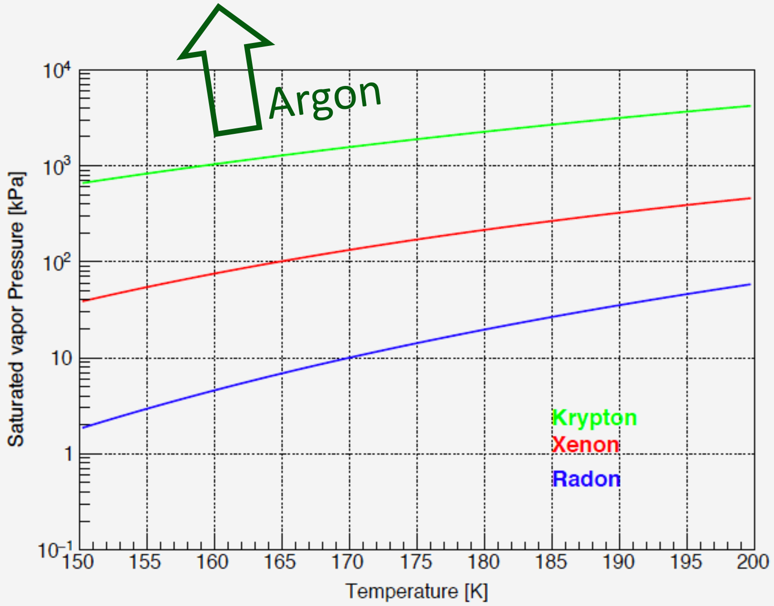
- $\boldsymbol{\alpha = 10.5}$ (@ -100 °C)
- **More volatile** Kr collected at the **top**
- Extract Kr-enriched Xe from the top (off-gas)
- Extract Kr-depleted Xe from the bottom
- Xe off-gas considered as loss

^{222}Rn

- $\boldsymbol{\alpha = 0.1}$ (@ -100 °C)
- **Less volatile** Rn collected at the **bottom**
- Trap Rn in the reboiler until decay ($t_{1/2} = 3.8$ d)
- Extract Rn-depleted Xe from the top

single distillation stage

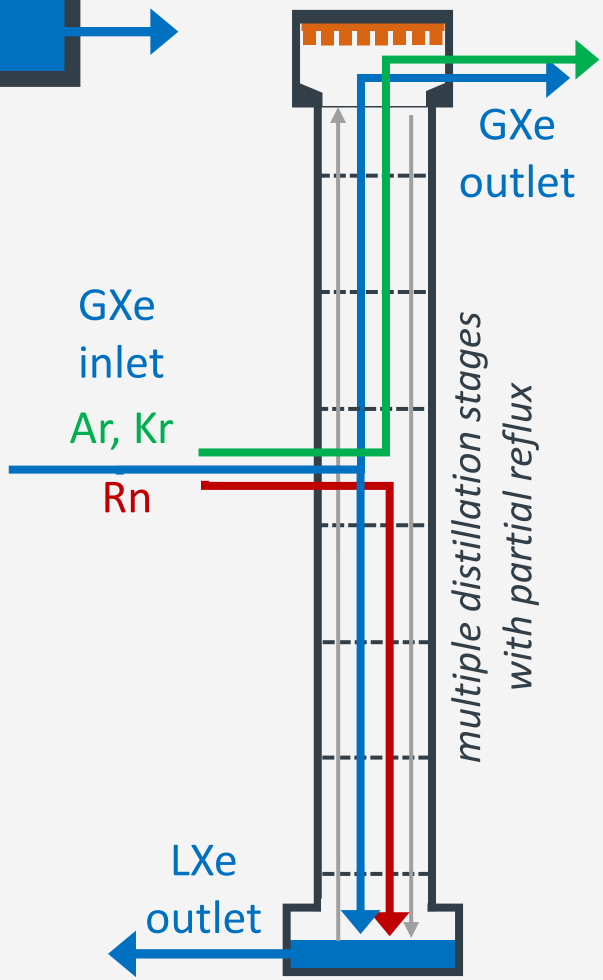




X. Cui et al. (PandaX Collaboration, JINST 16 (2021) P0704

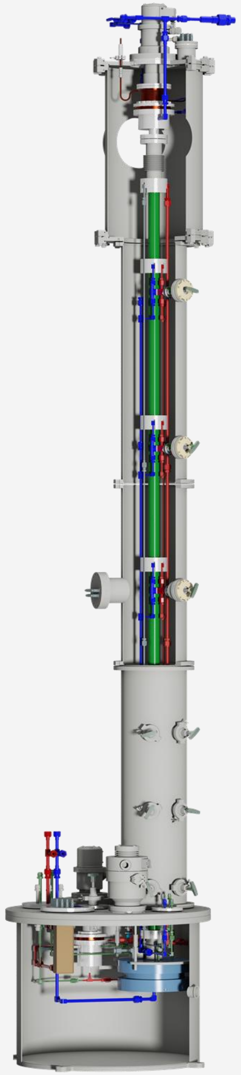
Transition probabilities of single noble gas atoms from gas to liquid and vice versa: **saturation vapor pressure**

condenser



reboiler

Operating krypton and radon removal systems @XENONnT



Krypton distillation

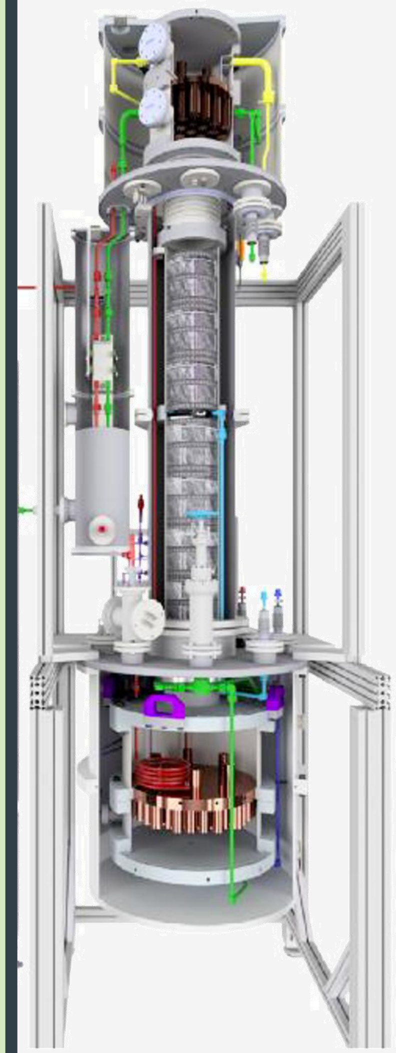
- Offline distillation demonstrated
  $^{nat}Kr/Xe < 0.026 \times 10^{-12}$ (26 ppq)
- Online distillation @XENON1T
  $^{nat}Kr/Xe < (360 \pm 60) \times 10^{-15}$ (ppq)
- **Offline + online distillation @XENONnT**
  $^{nat}Kr/Xe < (56 \pm 36) \times 10^{-15}$ (ppq)
  - Processed 6 t commercial xenon offline
  - Additional 3 weeks of online distillation
  - Further online distillation campaigns (Ar)

EPJ C 77 277 (2017), EPJ C77 358 (2017)
PTEP 053H01 (2022), EPJ C 82 1104 (2022)

- **Xe recovery:** 99% (1% off-gas)
- **1 day of Kr distillation generates ~ 0.7 kg off-gas**

- **Need for online distillation**
  - Kr can re-enter
  - ^{37}Ar calibrations

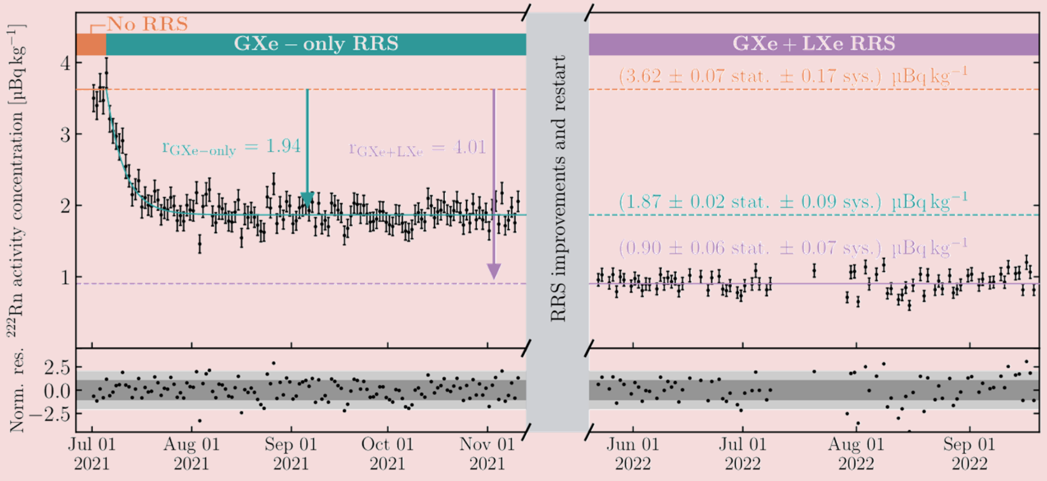
LowRad: $< 0.03 \times 10^{-12}$ (30 ppq)
DARWIN/XLZD: $< 0.1 \times 10^{-12}$ (100 ppq)



Radon distillation

- Continuous online distillation in various modes
- Reached radon-induced background rate comparable to the irreducible solar neutrino-induced background

- SR0 – GXe-only: 1.8 µBq/kg
- SR1 – **GXe + LXe: 0.9 µBq/kg** (200 slpm GXe + 25 slpm LXe)

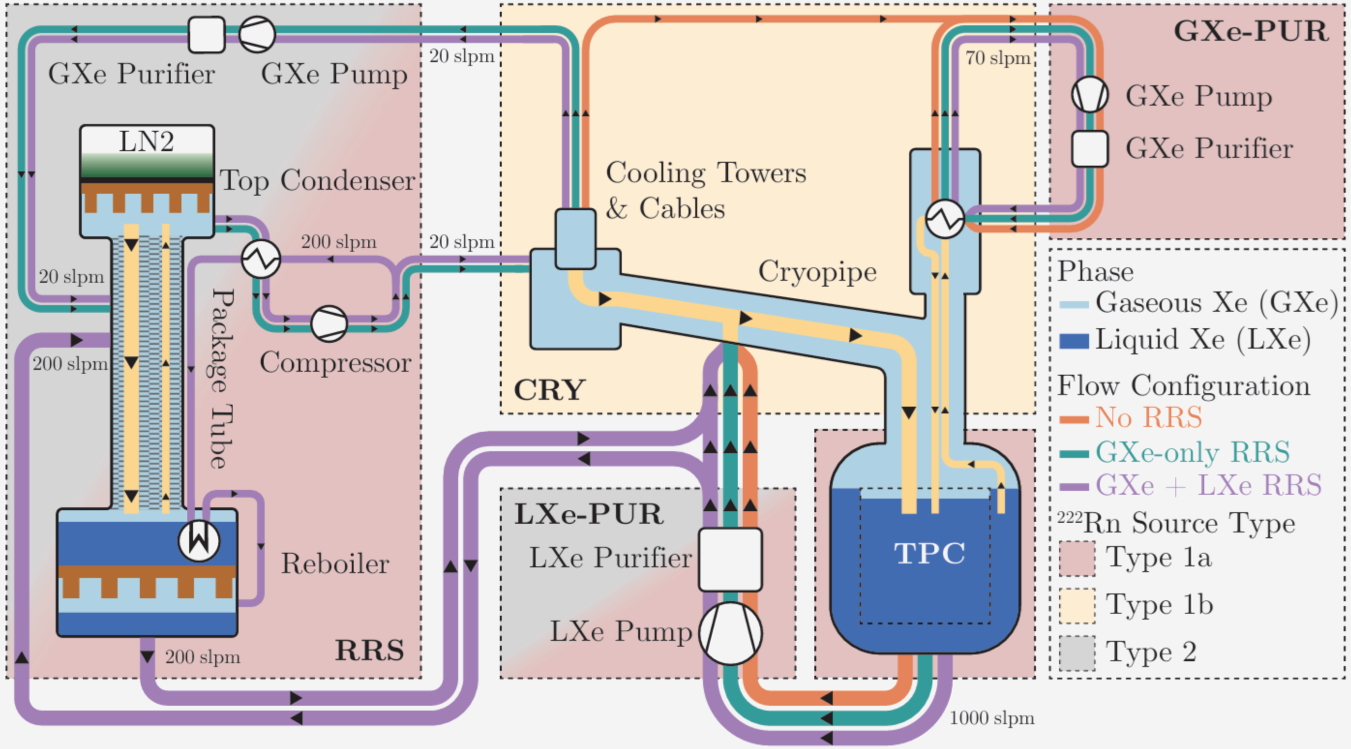


Radon Removal in XENONnT down to the Solar Neutrino Level
arXiv: 2502.04209

DARWIN/XLZD: ~0.1 µBq/kg (10^{-26})

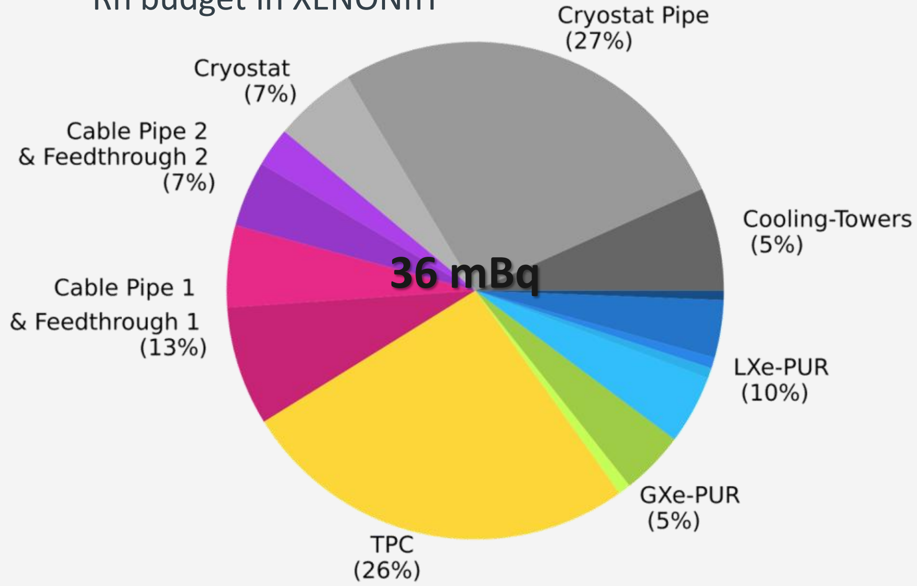
EPJC 82, 599 (2022)

Radon budget in XENONnT



Detailed description
Radon Removal in XENONnT down to the Solar Neutrino Level
arXiv: 2502.04209

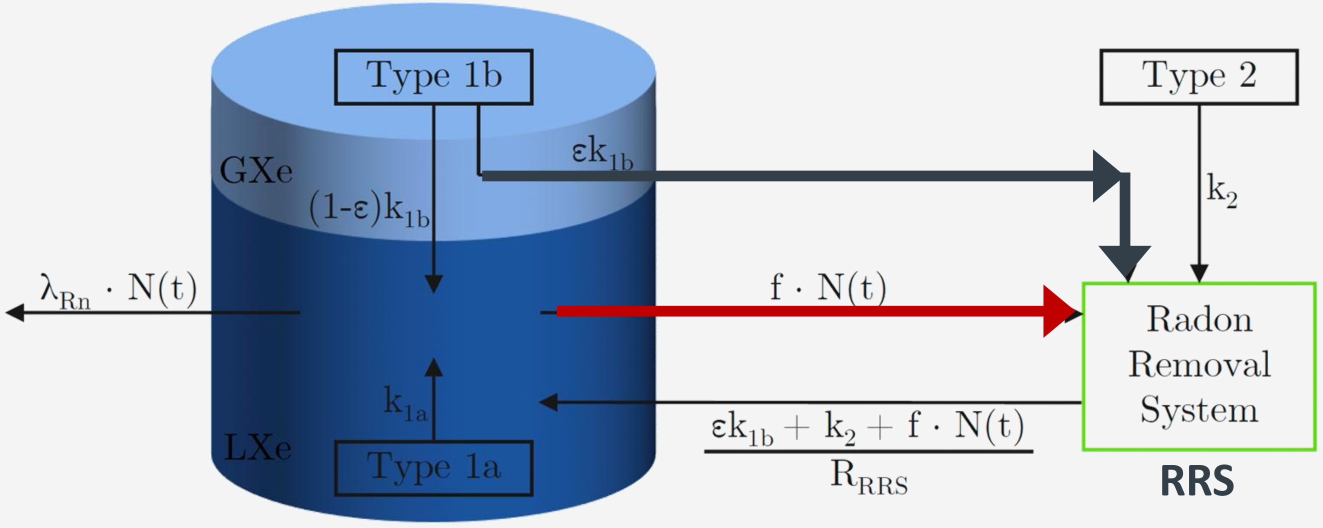
^{222}Rn budget in XENONnT



Different Rn source types:

- **Type 1b:** enters the **GXe phase** with a rate k_{1b} from cables or lines to the outside, can be extracted directly with a **fraction ε to the RRS**
- **Type 1a:** enters the **LXe phase** in the detector with a rate k_{1a}, can be extracted with a **low effective flow f to the RRS**
- **Type 2:** enters the **RRS** with a rate k_2 **before the LXe phase**

Reduction by continuous ^{222}Rn removal



Different Rn source types:

- **Type 1b:** enters the **GXe phase** with a rate k_{1b} from cables or lines to the outside, can be extracted directly with a **fraction ε to the RRS**
- **Type 1a:** enters the **LXe phase** in the detector with a rate k_{1a}, can be extracted with a **low effective flow f to the RRS**
- **Type 2:** enters the **RRS** with a rate k_2 **before the LXe phase**

$$r(R_{\mathrm{RRS}} \to \infty, f, \epsilon) = \underbrace{\frac{\lambda_{\mathrm{Rn}} + f}{\lambda_{\mathrm{Rn}}}} \cdot \underbrace{\frac{k_{\mathrm{tot}}}{k_{1\mathrm{a}} + (1-\epsilon)k_{1\mathrm{b}}}}$$

$\approx \mathbf{2} \cdot \mathbf{2} = \mathbf{4}$ for a XENONnT at a flow of ≈ 80 kg/h

LXe extraction reduction factor with total LXe exchange time T: $r_{\mathrm{LXe}} \approx 1 + {}^{\tau_{\mathrm{Rn}}}/_{T} \approx 2$ to 4

GXe extraction reduction factor, typically $r_{\mathrm{GXe}} \approx 2$ (XENONnT)

Online distillation:
E. Aprile et al. (XENON Collab.)
Prog. Theo. Exp. Phys. 5 (2022) 053H01

Radon removal system @XENONnT

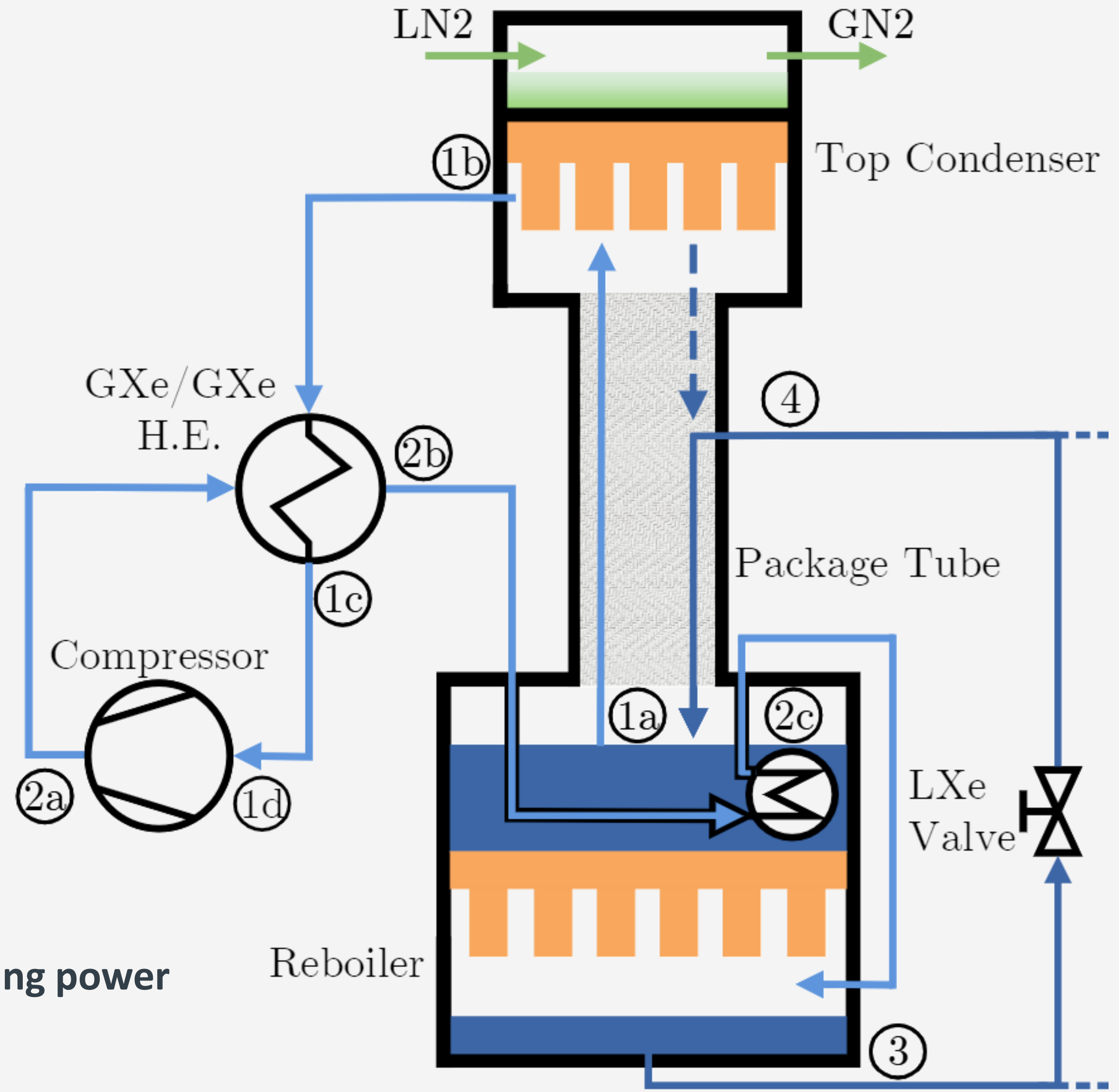
Design parameters:

- **Target flow:** 72 kg/h (200 slpm)
- Requires 1 kW cooling power at top
- LXe inlet and outlet require 2 kW cooling power

Thermodynamic concept:

- **Clausius-Rankine cycle** with phase changing medium xenon
- Reboiler acts as **heat exchanger** to liquefy Rn-depleted GXe with the stored Rn-enriched LXe
- Compressor acts as **heat-pump**

- Reduce required external cooling power from **3 kW to 1 kW**
- Drastically **reduce nitrogen consumption** and **electrical heating power**



Radon-free compressor
JINST 16 P09011 (2022), based on EPJ C78 604 (2018)

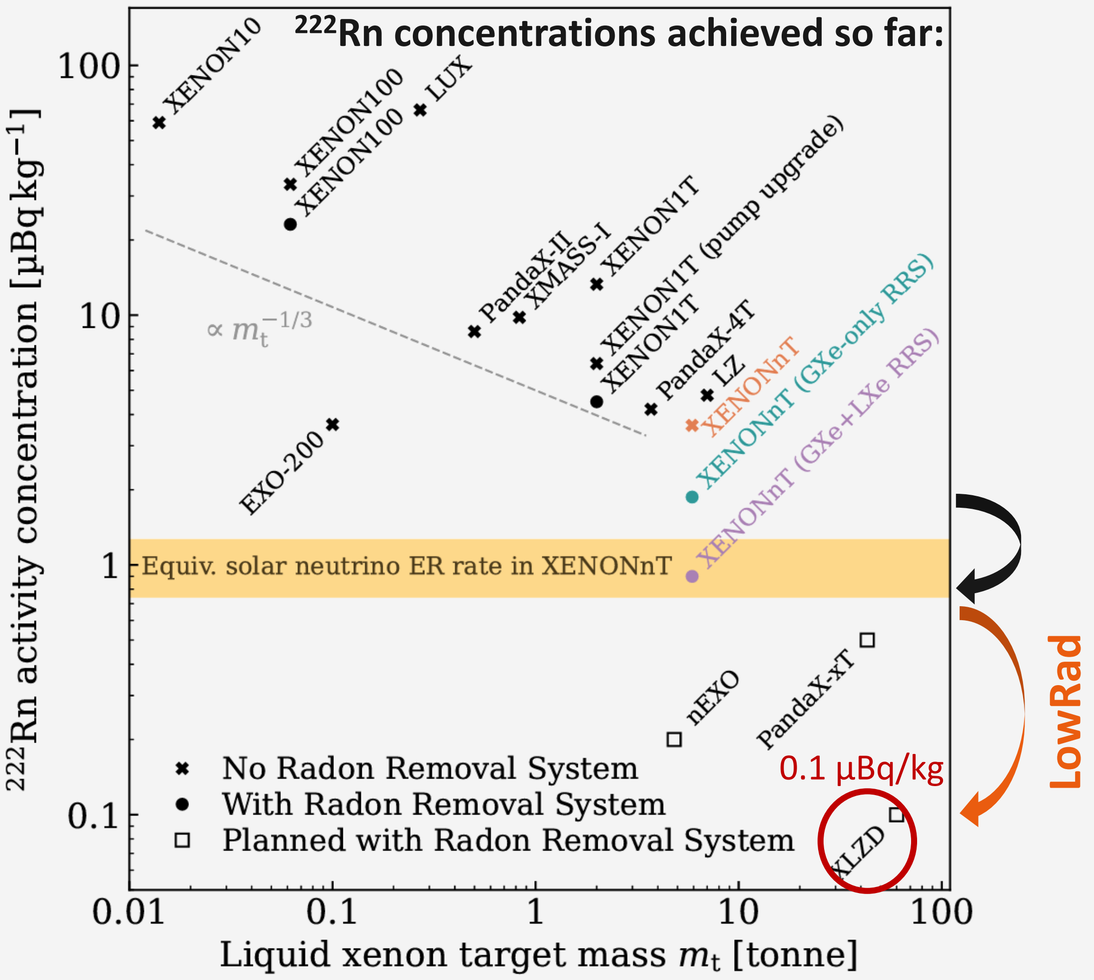
Radon-free heat exchangers
JINST 17 P05037 (2022)

Low radon and low internal radioactivity – LowRad

ERC Advanced Grant LowRad of C. Weinheimer:

- **Develop technologies** for:
  - Next-generation experiments with around **75 t of LXe**
  - Continuous/**online ^{85}Kr removal** (30 ppq ^{nat}Kr)
  - Another **factor 10** in **^{222}Rn reduction** (0.1 µBq/kg) together with other Rn mitigation techniques
- R&D for novel purification methods
- Methods for physics searches/analyses
- Pave the way for an **all-in-one** purification & distillation system
- Complete **purification & distillation demonstrator**

Reach: “less than 1 Radon atom in 100 mol of xenon”



Rn background rate 10 times smaller than solar neutrino contribution

Key aspects of the LowRad project

How to purify 75 t of Xe from Rn in ≤ 2d ?

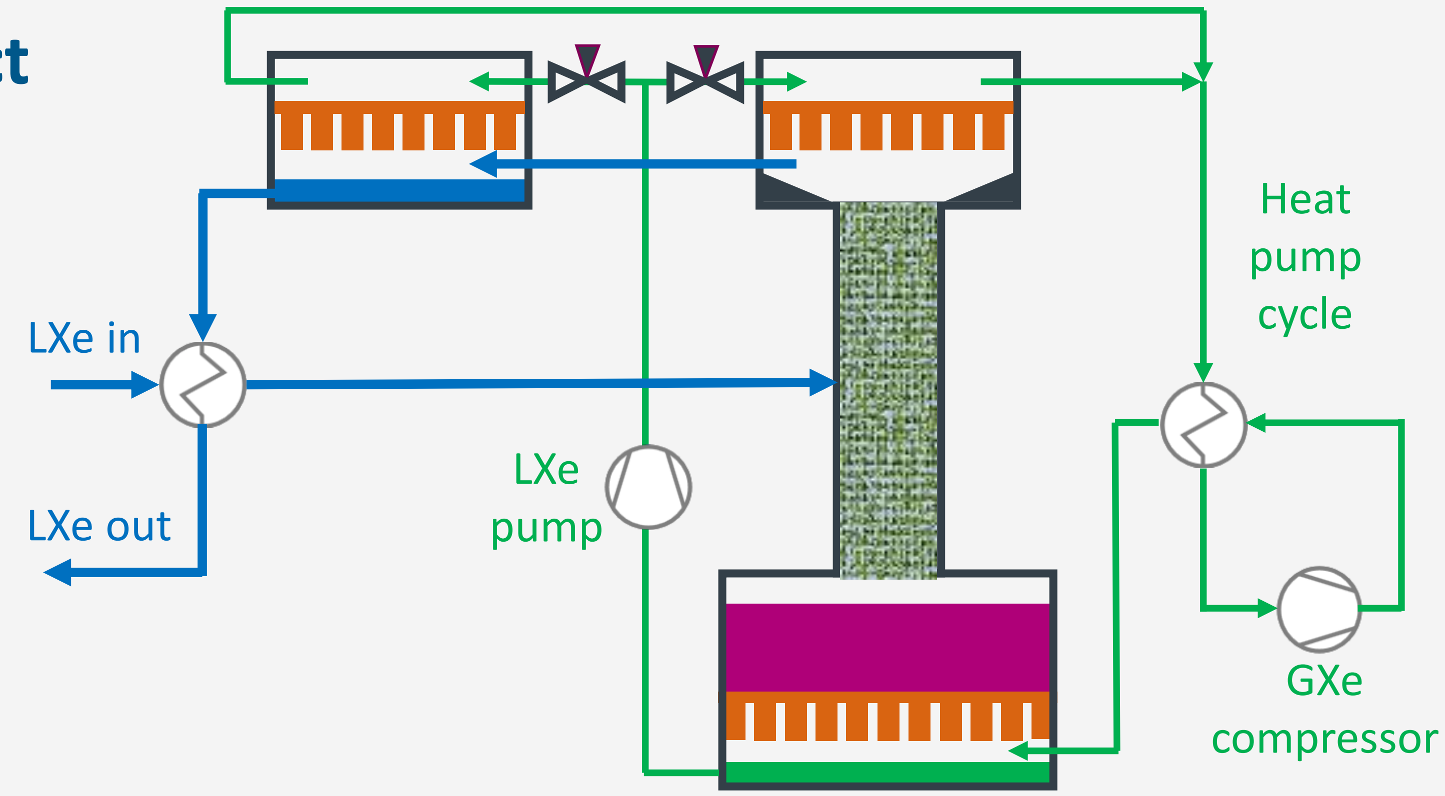
- **Full heat pump** to achieve enormous cooling
  throughput: **75 kg/h (LowRad demonstrator)**
  1.7 t/h (final system)

Demonstrator

- Radon-free heat exchangers
- 2^{nd} Xe **heat pump** cycle
- With online **Rn decay monitor**

Final system

- Should be integrated with **purification system** for removal of electronegative impurities
- With **online Kr removal system**
- Installed in a **water shield** to avoid Xe activation



R&D currently focusing on

- **Kr-concentrator** for an online distillation system
- Demonstration of a **Xe heat pump** concept
- **Rn-decay detectors** for efficiency monitoring
- Purification system (purity monitor & clean pumps)

Online Krypton distillation system

Challenges of the XENON1T/nT system

- Initially **not designed for continuous online distillation**
- Limited to gas flows of up to 18 slpm = 6.5 kg/h
- Planned for an **off-gas fraction of 1%**
- **Replacement** of the **off-gas bottle** after ~60 days of distillation

Online distillation for DARWIN/XLZD

Assuming a runtime of 5 years for a 75 t detector

distillation speed

off-gas 1 fraction

$$5\,\mathrm{yr} \times 8.3\,\mathrm{slpm} \times \frac{75\,\mathrm{t}}{8.6\,\mathrm{t}} \times 10^{-2}$$ ⇒ 6 kg/d off-gas 1

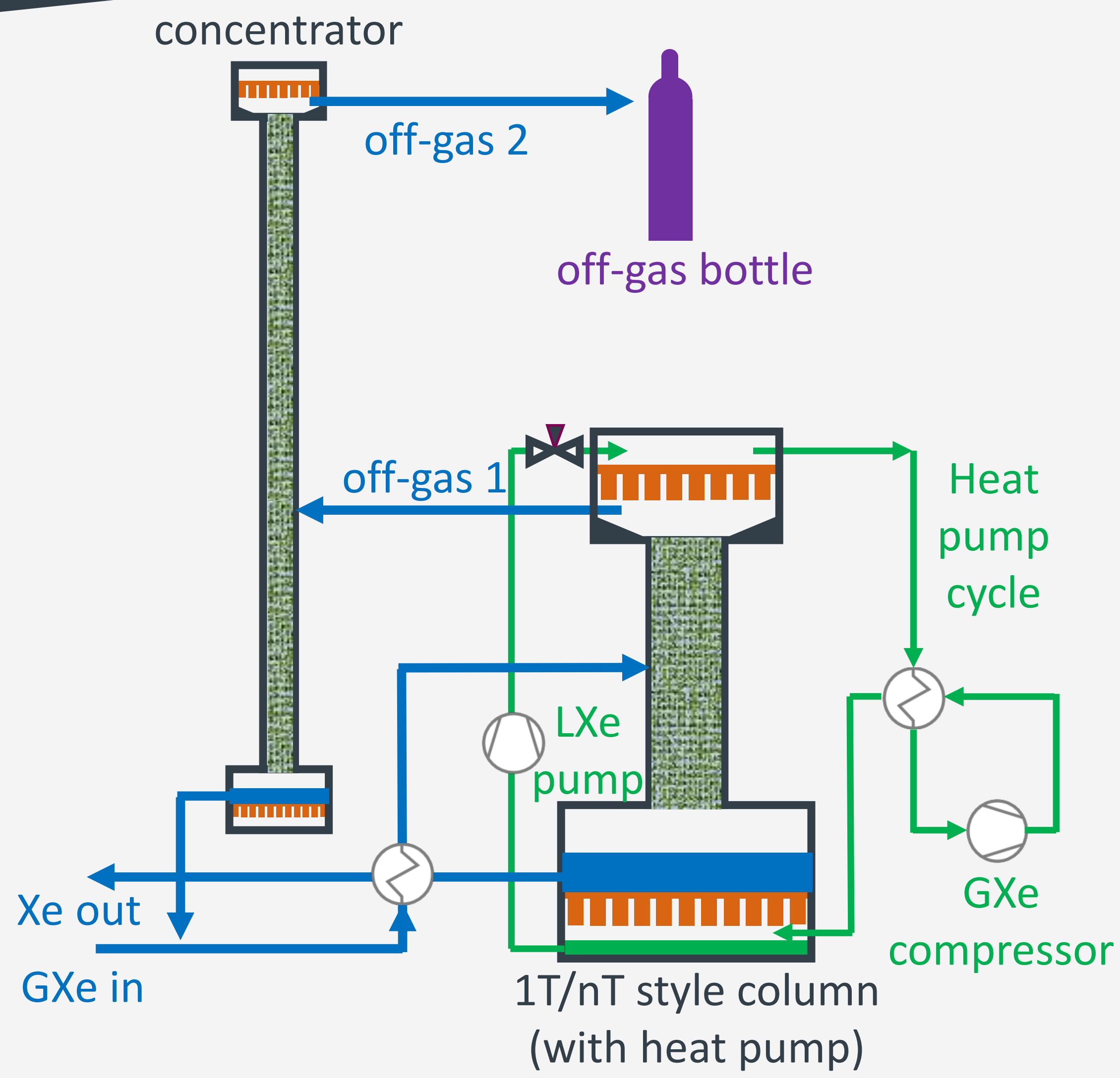
runtime

detector mass scaling

$$\times\, 10^{-3} = 11.4\,\mathrm{kg}$$ ⇒ 6 g/d off-gas 2

off-gas 2 fraction

⇒ Can be easily compensated

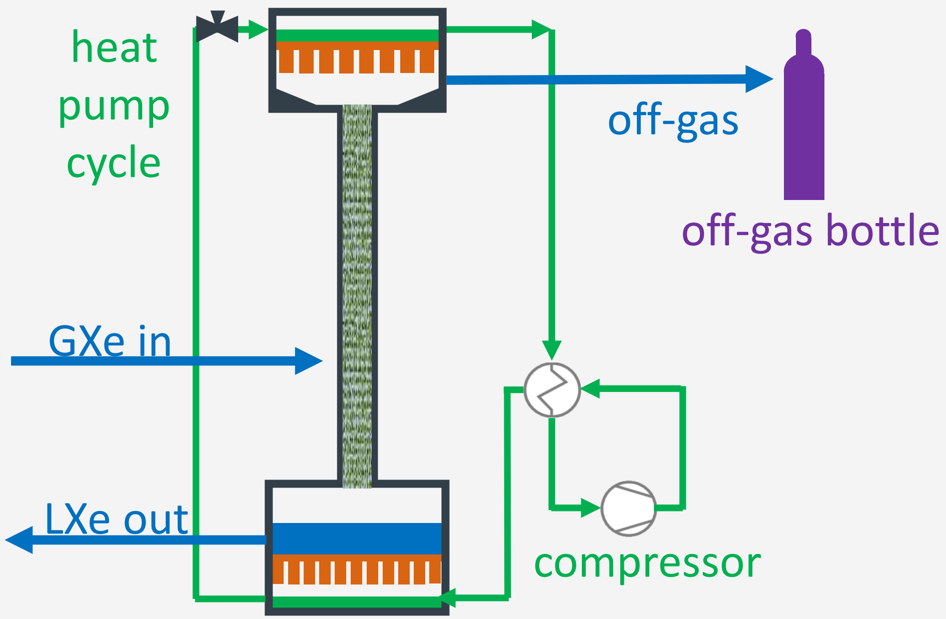


LowRad concentrator prototype

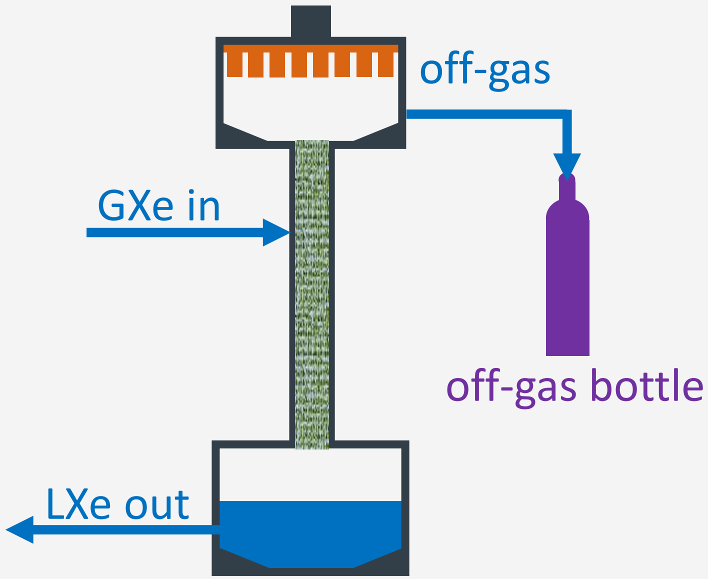
I: Krypton distillation column
- Design as a tool – “as simple as possible”
- Demonstration of **high separation** and the 10^{-3} **off-gas fraction** for **low flows** (< 5 slpm)

II: Xe heat pump
- Design and test of a **heat pump cycle** using a suitable process gas (design for Xe)

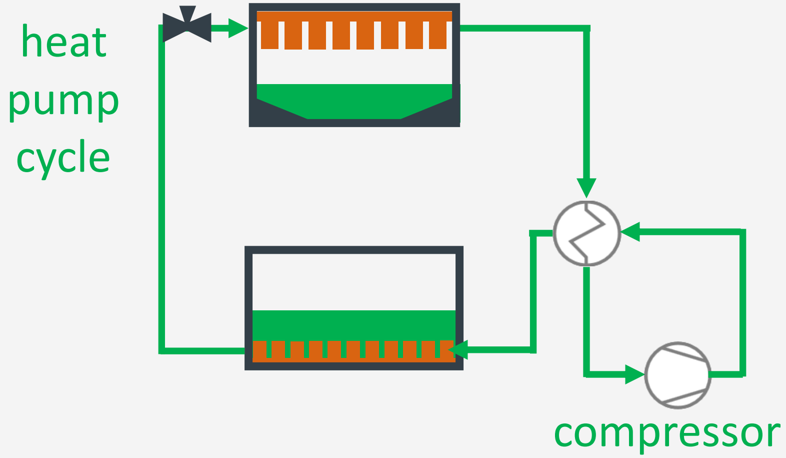
Krypton concentrator prototype



I: distillation column



II: Xe heat pump



I: Krypton distillation column

Development of a McCabe-Thiele calculator

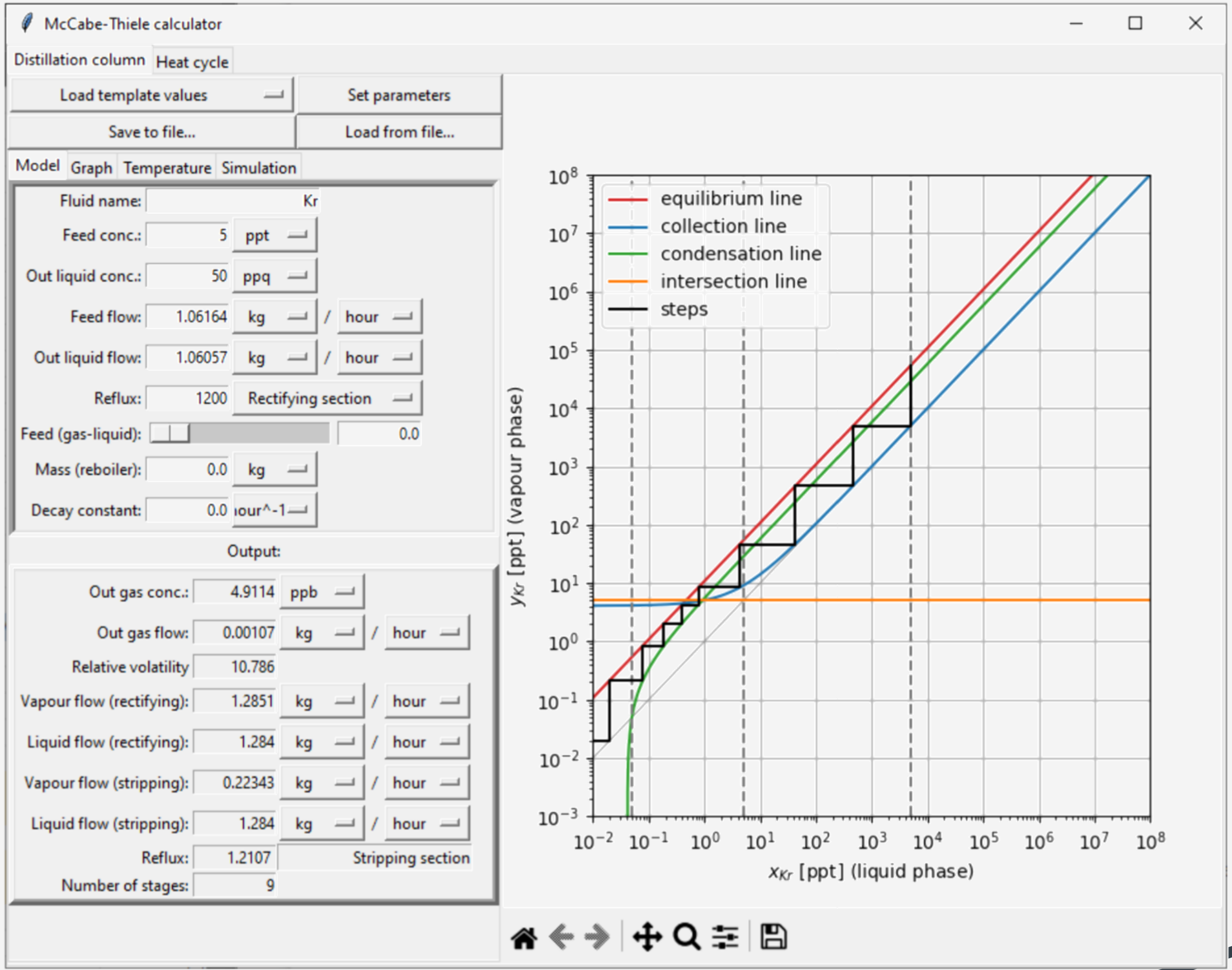
- Versatile McCabe-Thiele calculator
- Estimation for **required number of distillation** stages and **expected performance**
- Arbitrary selection of conditions
- Integration of TESPy and CoolProb tools

Requirements of the column

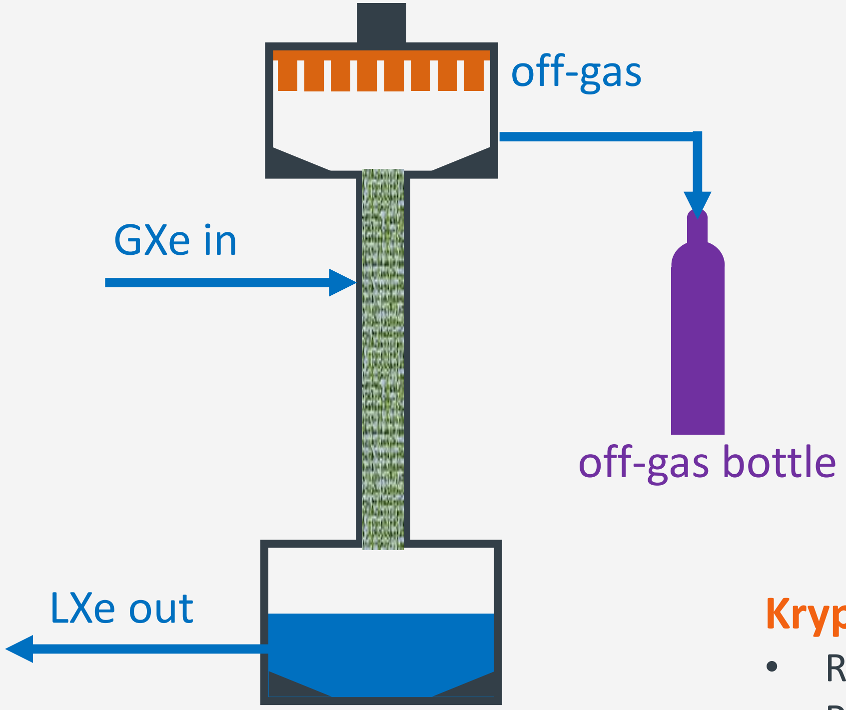
- 1 kg/h (3 slpm) xenon flow
- 0.1 % offgas flow
- In: ^{nat}Kr/Xe of 5×10^{-12} (ppt)
- Out: ^{nat}Kr/Xe of 50×10^{-15} (ppq)

Theoretical calculation

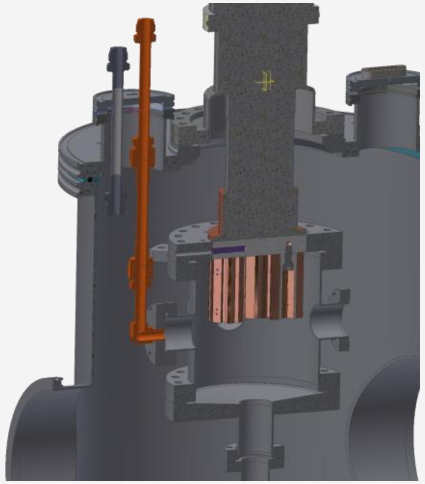
- Offgas: ^{nat}Kr/Xe of 10^{-9} (ppb)
- Requires 9 distillation stages
- Reflux of 1200 (internal circulation)
- Requires 40 W of additional cooling power



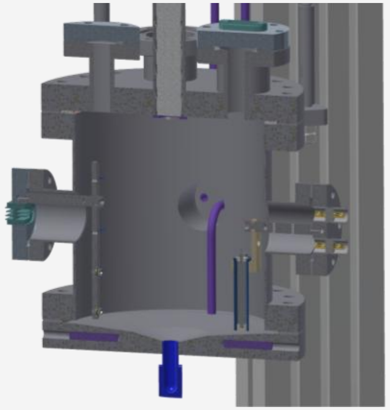
I: Krypton distillation column

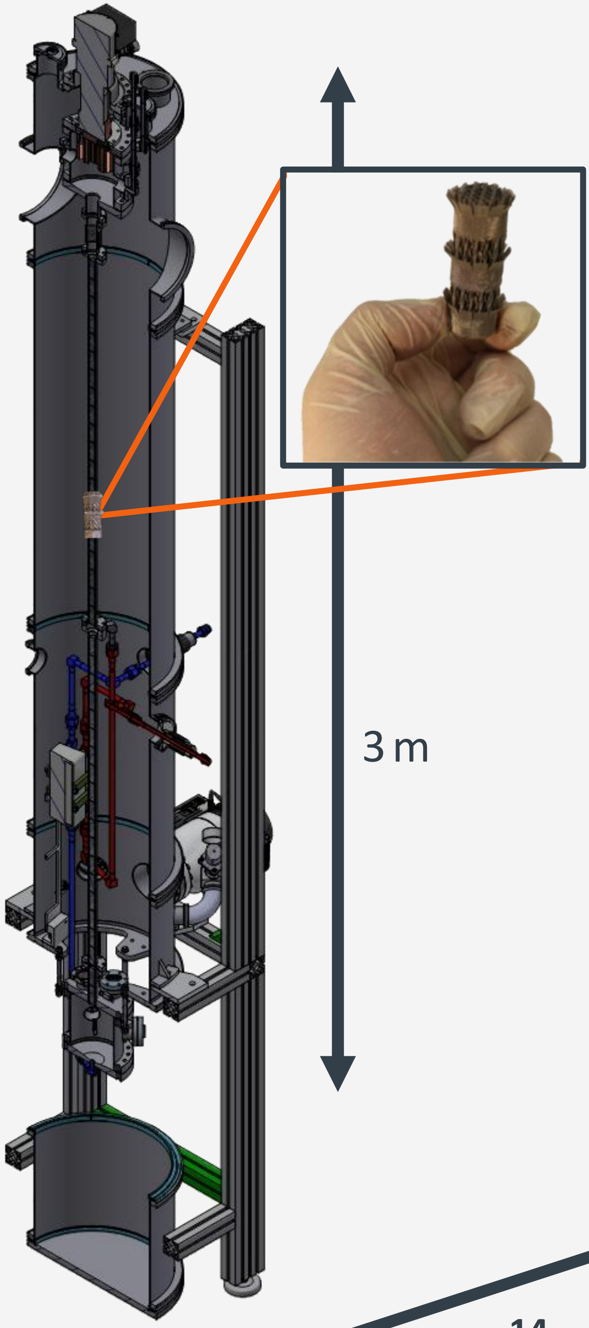


Top condenser



Bottom reboiler





Inlet: 1 kg/h (3 slpm)
Reflux: 1200
Theoretical stages: 9

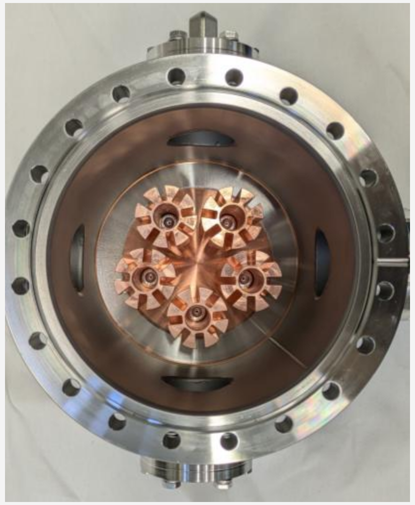
Krypton distillation column

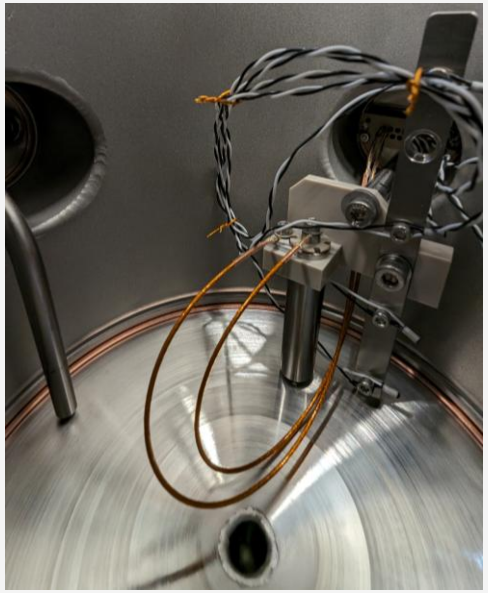
- Requires 40 W of additional cooling power
- Package height of 2.2 m (extension possible)
- Using a 2 cm diameter package material (type EX, Sulzer AG)
- Modular design to switch to heat pump concept (re-evaluate performance)
- Characterize **separation** and show $\mathbf{10^{-3}}$ **off-gas fraction** for **low flows** (few slpm)

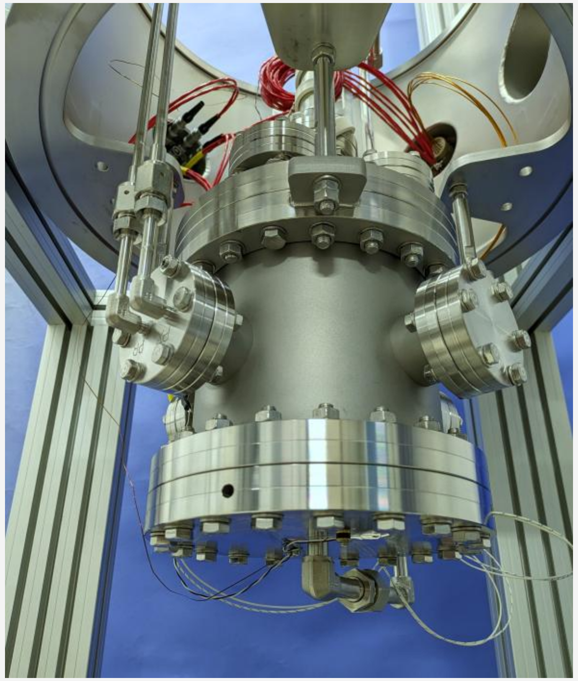
I: Krypton distillation column

- Construction of the system started in April 2024
- **August**: Installed and connected to main gas system
- **September**: Baking and extensive cleaning
- **October**: First xenon filling and thermodynamic tests
- **Currently**: Characterization of the column performance
  - Test various inlet/outlet flows vs off-gas fraction
  - Demonstrate the 10^{-3} off-gas fraction
  - …

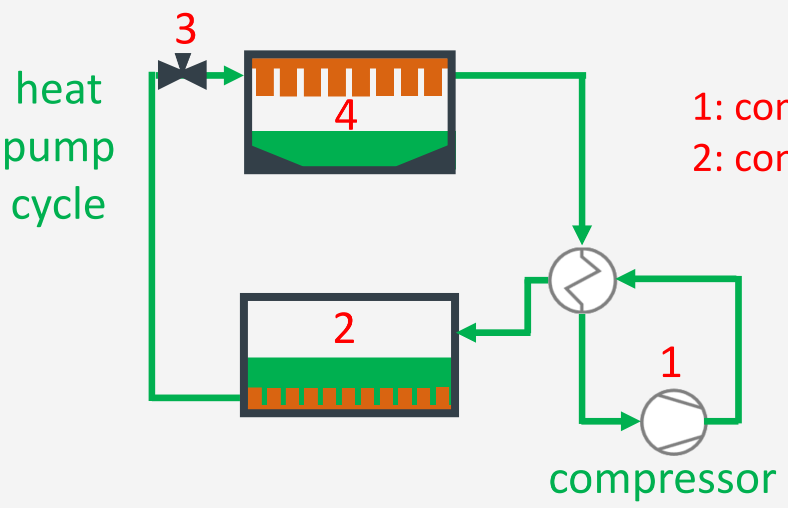
3 m



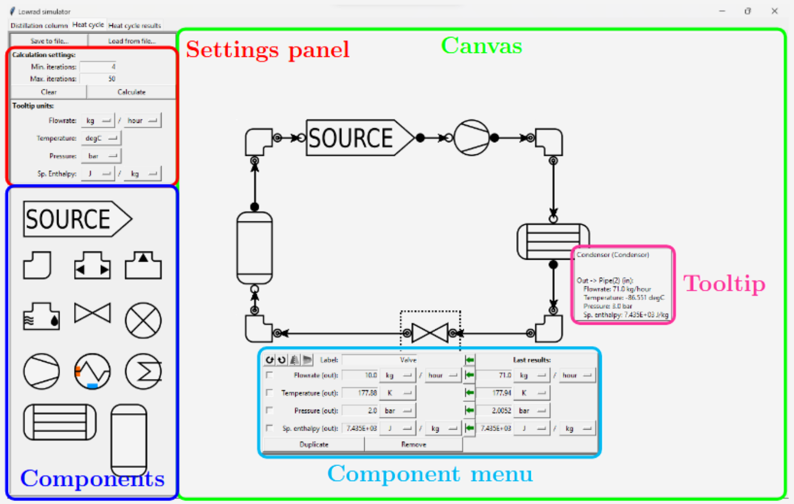


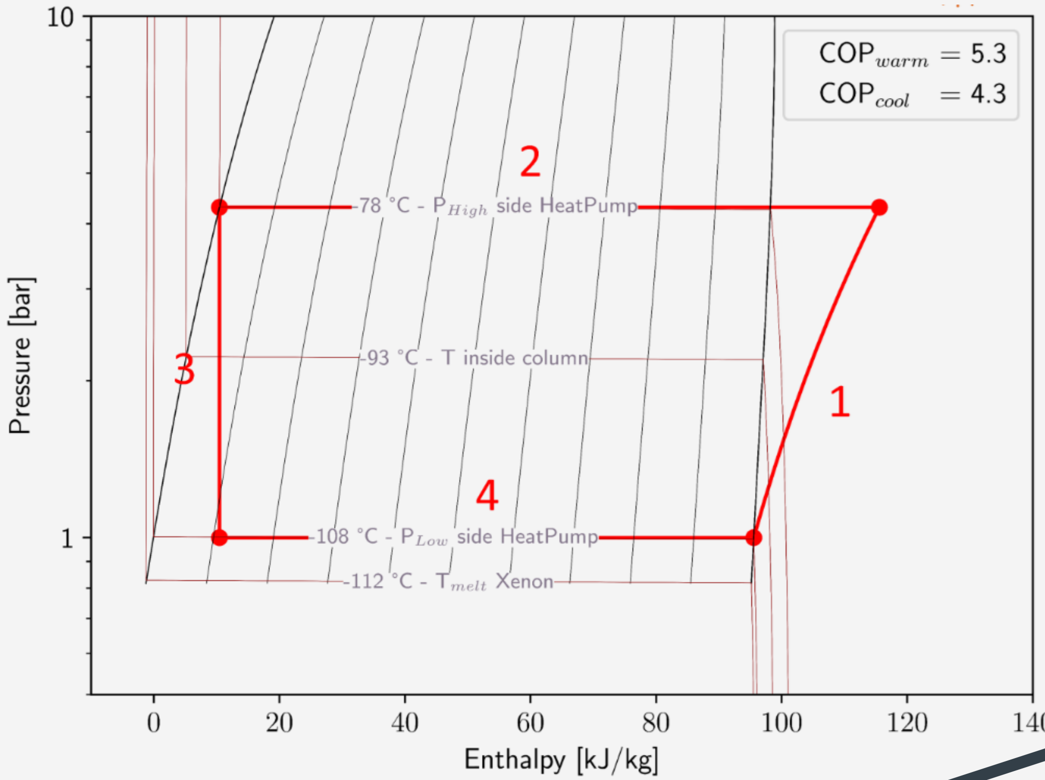


II: Xe heat pump



1: compression
2: condensation
3: expansion
4: evaporation



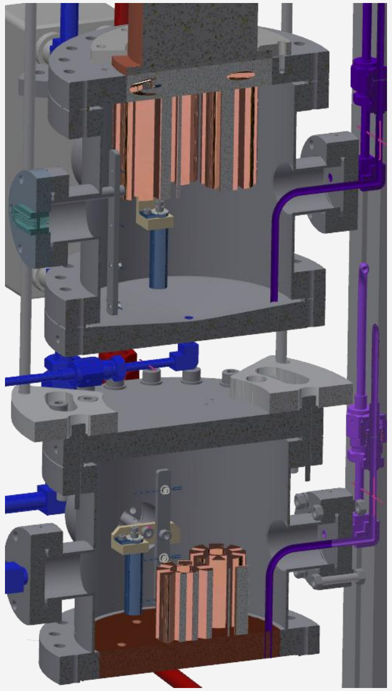


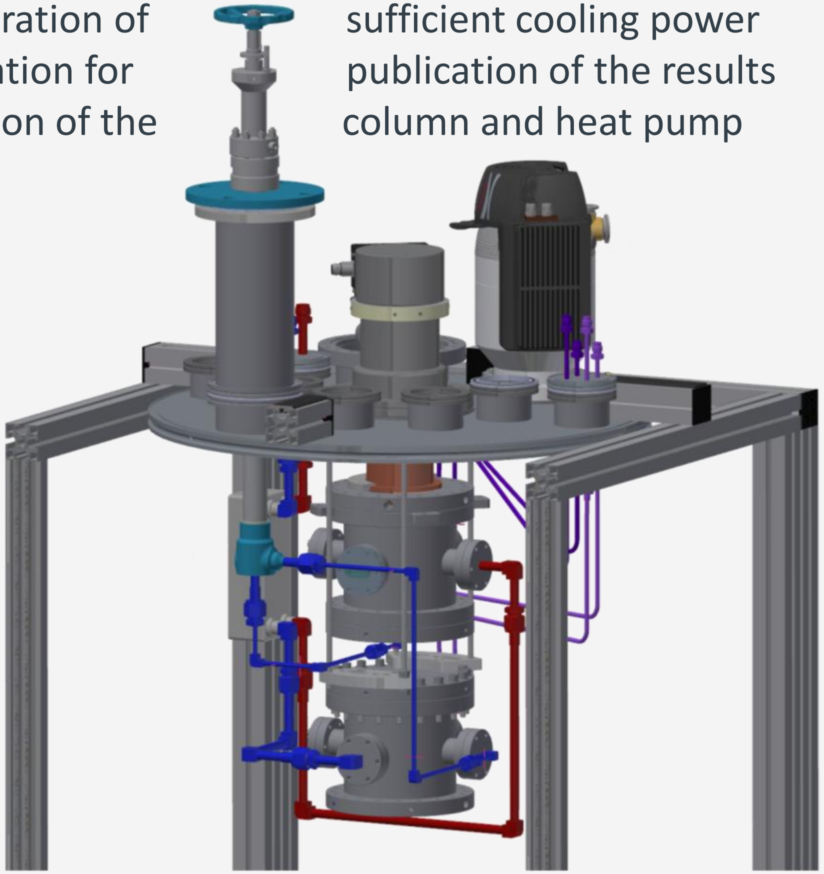
Design of a heat pump cycle

- Investigating several process gases including **Xe**
- Need to supply the required heating and cooling power at reboiler and condenser (roughly 10 W per 1 slpm)
- Design using custom numerical calculator for heat pump concepts/heat cycles

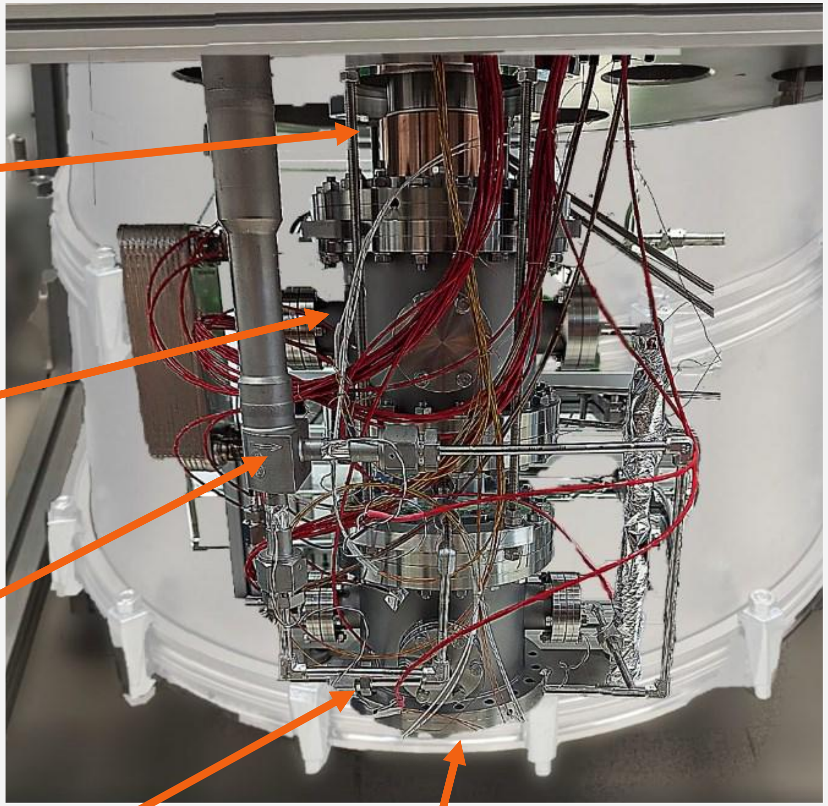
II: Xe heat pump

- Construction of the system started in April 2024
- **September:** First LXe filling and circulation
- **October:** Further tests of the Xe circulation (with LXe)
- **December:** Test stability and cooling performance
- **January:** Demonstration of sufficient cooling power
- **Currently:** Preparation for publication of the results
- **Future:** Combination of the column and heat pump





1: compression 3: expansion
2: condensation 4: evaporation



cold head

condenser vessel
2

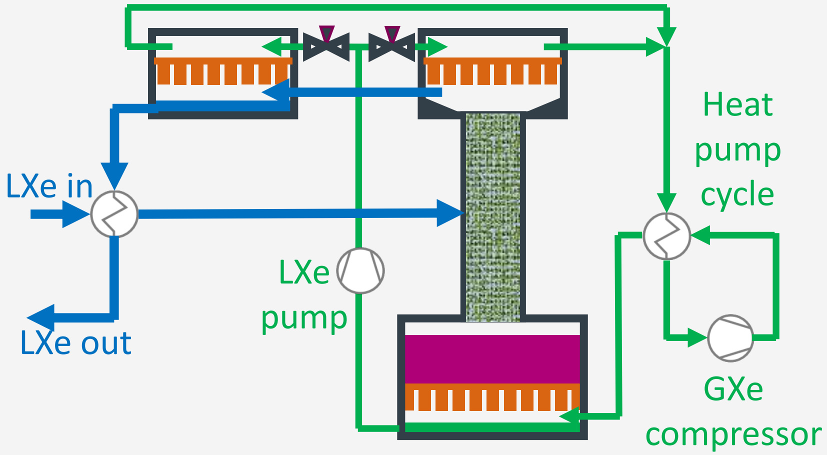
expansion valve
3

reboiler vessel
4

heating elements (simulating a column condenser)

Conclusion – The LowRad project

- **Develop technologies** for:
  - Continuous/**online ^{85}Kr removal** (30 ppq ^{nat}Kr)
  - Another **factor 10** in **^{222}Rn reduction** (0.1 µBq/kg)
  - Online **concentration monitoring**
- Design of a **purification & distillation demonstrator (75 kg/h)** with heat pump concept ongoing, construction in end of 2025
- **Other LowRad R&D at Münster** not shown today:
  - Kr- and Rn-decay monitoring systems
  - Ultra clean gas and liquid xenon pumps
  - Xenon gas purity monitoring systems



Krypton concentrator prototype

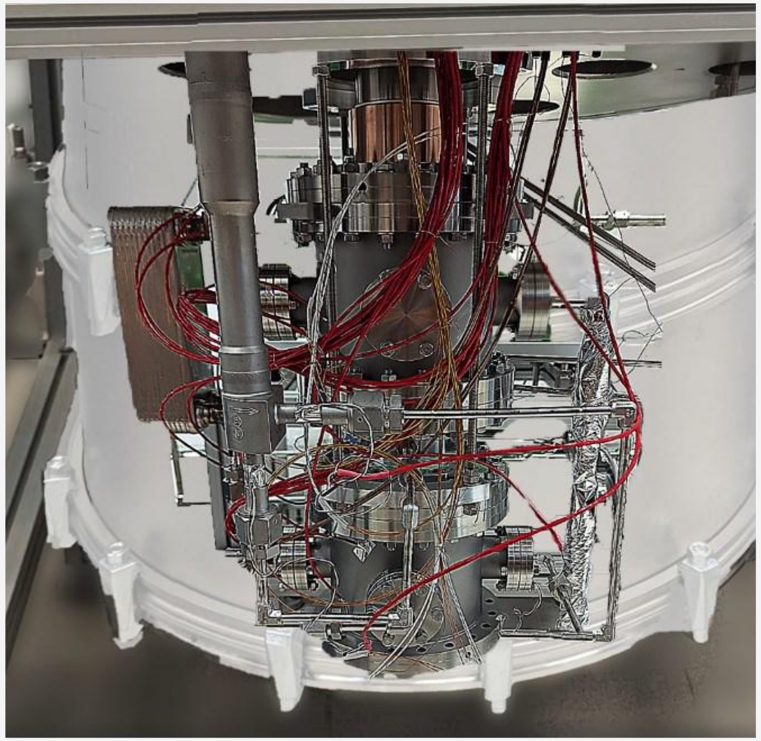
I: Krypton distillation column

→ Performance measurements @design speed

II: Xe heat pump

→ Demonstrated sufficient cooling power

→ Installation at the distillation column



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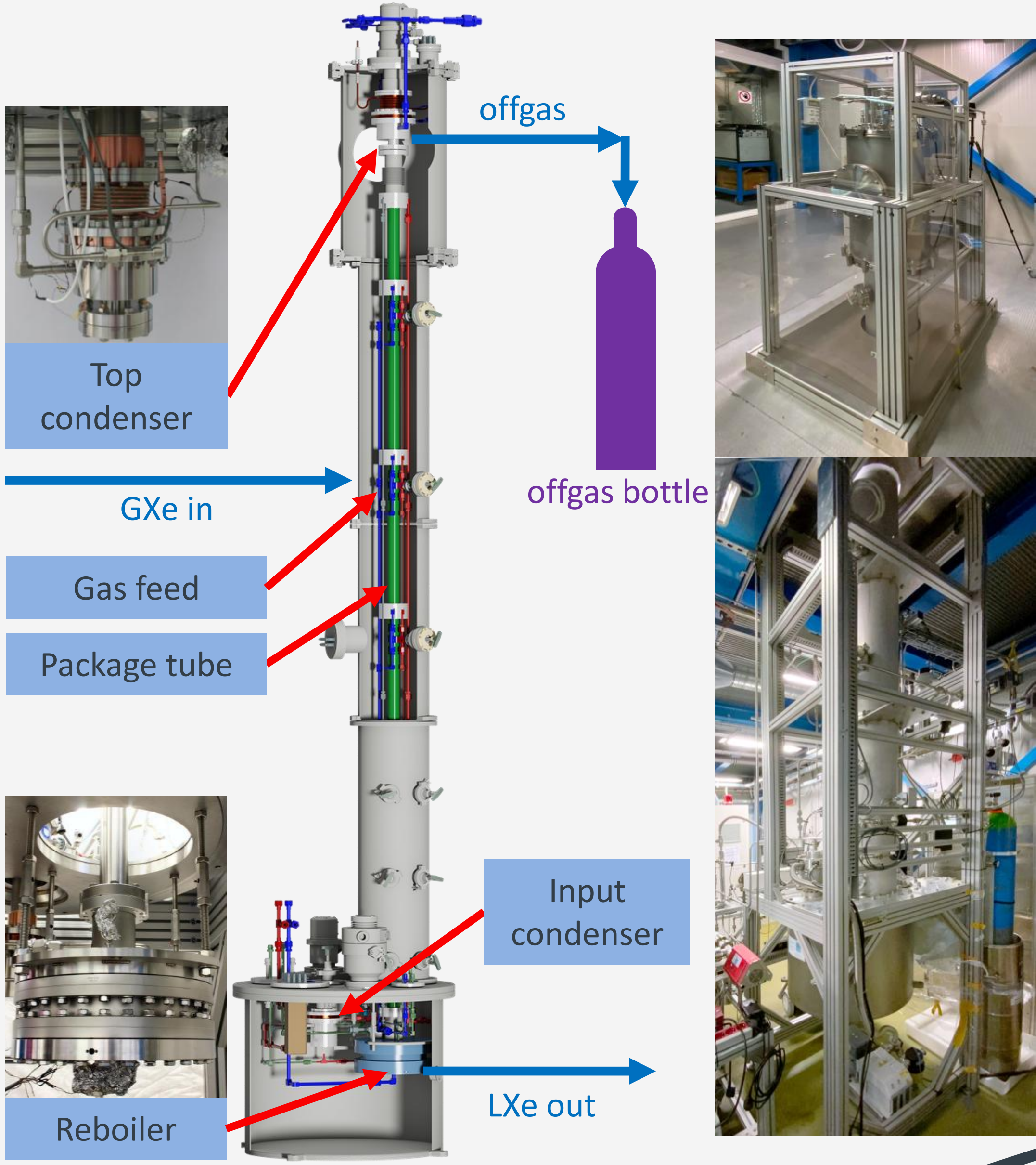
DFG
erc

Krypton distillation column for XENON1T/nT

Phase-2 DST column (design parameter | @XENON1T)

- Feeding flow rate: 8.3 slpm (3 kg/h)
  Thermodynamically stable up to 18 slpm (6.5 kg/h)
- Separation factor: 10^4 - 10^5
  Measured separation of $6.4^{+1.9}_{-1.4} \times 10^5$ at 8.3 slpm
- Kr removal: ^{nat}Kr/Xe $< 0.2 \times 10^{-12}$ (0.2 ppt)
  ^{nat}Kr/Xe $< 0.026 \times 10^{-12}$ (26 ppq) (Phase-1) (multiple cycles)
  ^{nat}Kr/Xe $< 0.048 \times 10^{-12}$ (48 ppq) (Phase-2) (single cycle)
- Xe recovery: 99% (1% offgas)
- Performance sufficient for XENONnT and DARWIN/XLZD

PhD S. Rosendahl (2015)
Eur. Phys. J. C 77 (2017) 275
PhD M. Murra (2019)
PTEP 5, 053H01 (2022)



Eur. Phys. J. C 77 (2017) 275

Online krypton removal for XENON1T

Requirements for ^{85}Kr removal system:

- Needs to be **removed once**
- Kr can re-enter via (tiny) air leaks

Reality check @XENON1T:

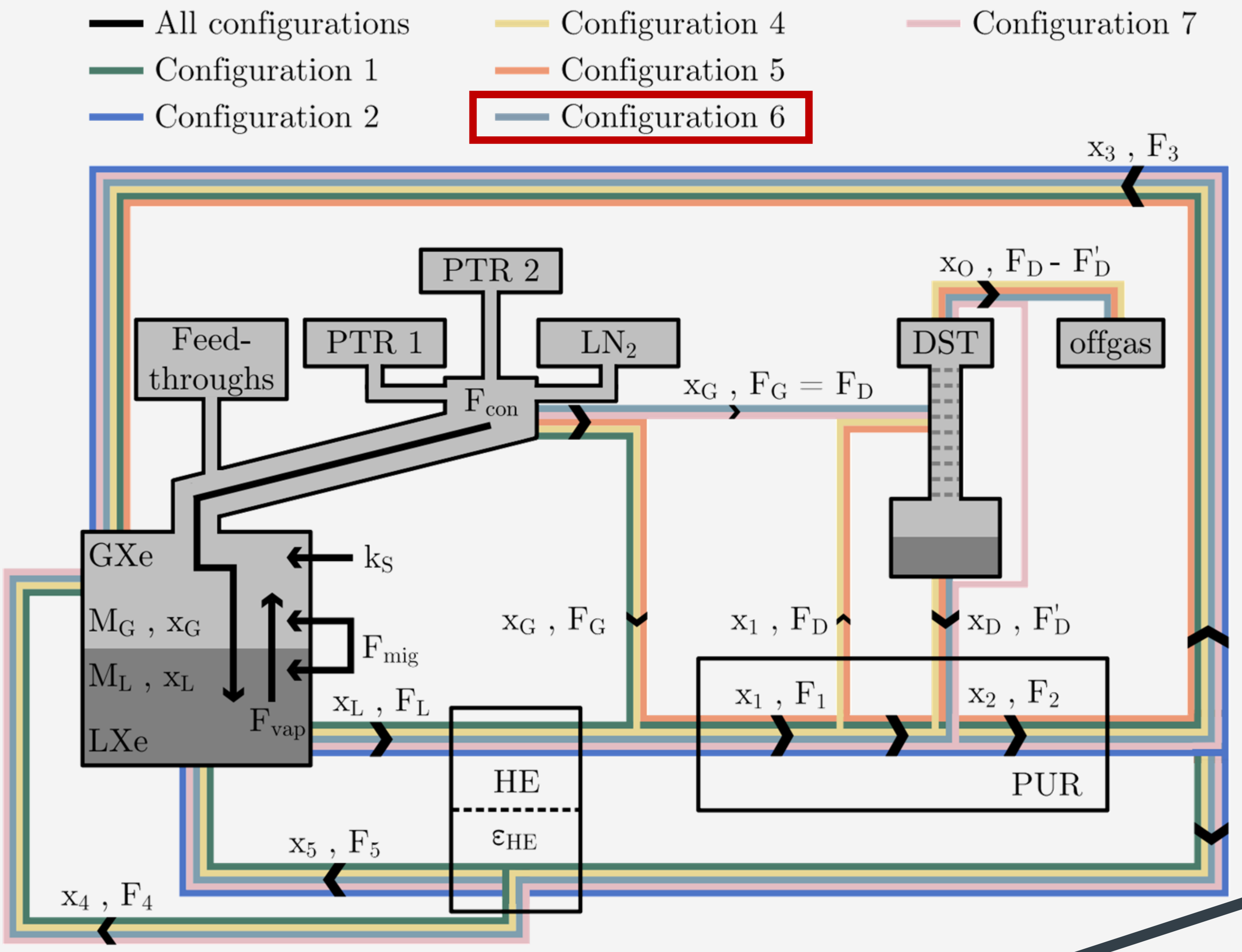
- Estimated 3-4 weeks distillation for 3.2 t Xe
- **Filled TPC without distillation** to test functionality

[a few weeks of commissioning]

- **Need** to apply **^{85}Kr removal**! But **without re-filling**!
- Development of the **online Kr removal method**
  Idea: S. Lindemann, M. Murra, G. Plante
  PTEP 5, 053H01 (2022)

Online Kr removal method

- Make use of the TPC as single stage DST
- Remove Kr from GXe, disturb equilibrium
- Kr migrates from LXe to GXe
- 1% offgas (0.85 kg/d | 6 kg/week), $\tau_{\text{eff, Kr}} = 6$ d

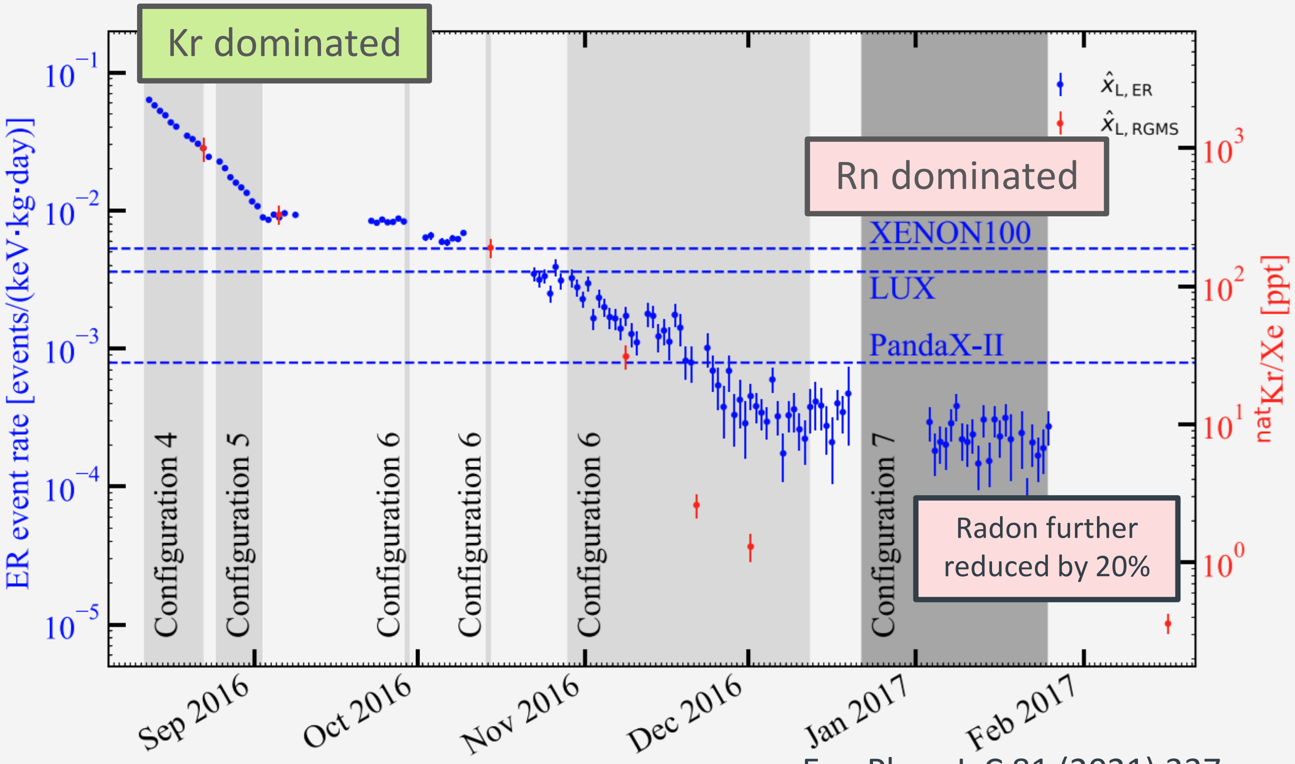


PTEP 5, 053H01 (2022)

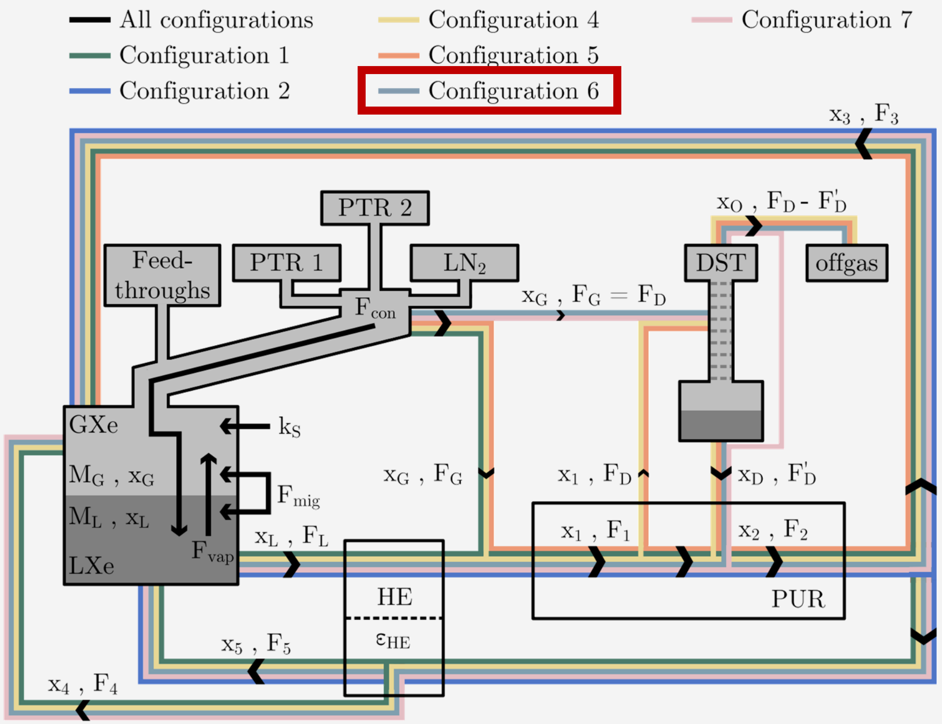
Online krypton removal for XENON1T

- Online Kr removal in configuration 4/5/6
- Reached a Kr conc. of ^{nat}**Kr/Xe** $< (360 \pm 60) \times 10^{-15}$ **(ppq)**

- Online Rn removal in configuration 7
- 20% reduction, no Kr conc. increase observed



Eur. Phys. J. C 81 (2021) 337



Event Rate in TPC (**ER**): Data give direct „online" insight to innermost 700 kg of LXe (FV)

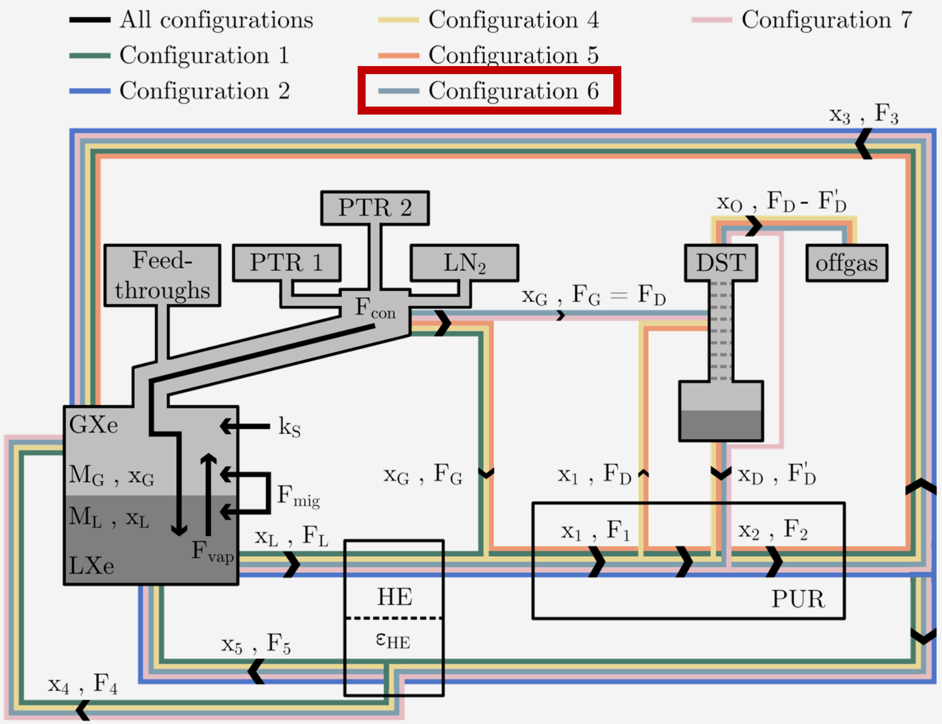
Rare Gas Mass Spectrometer (**RGMS**): Samples extracted from LXe for off-site analysis at MPIK Heidelberg

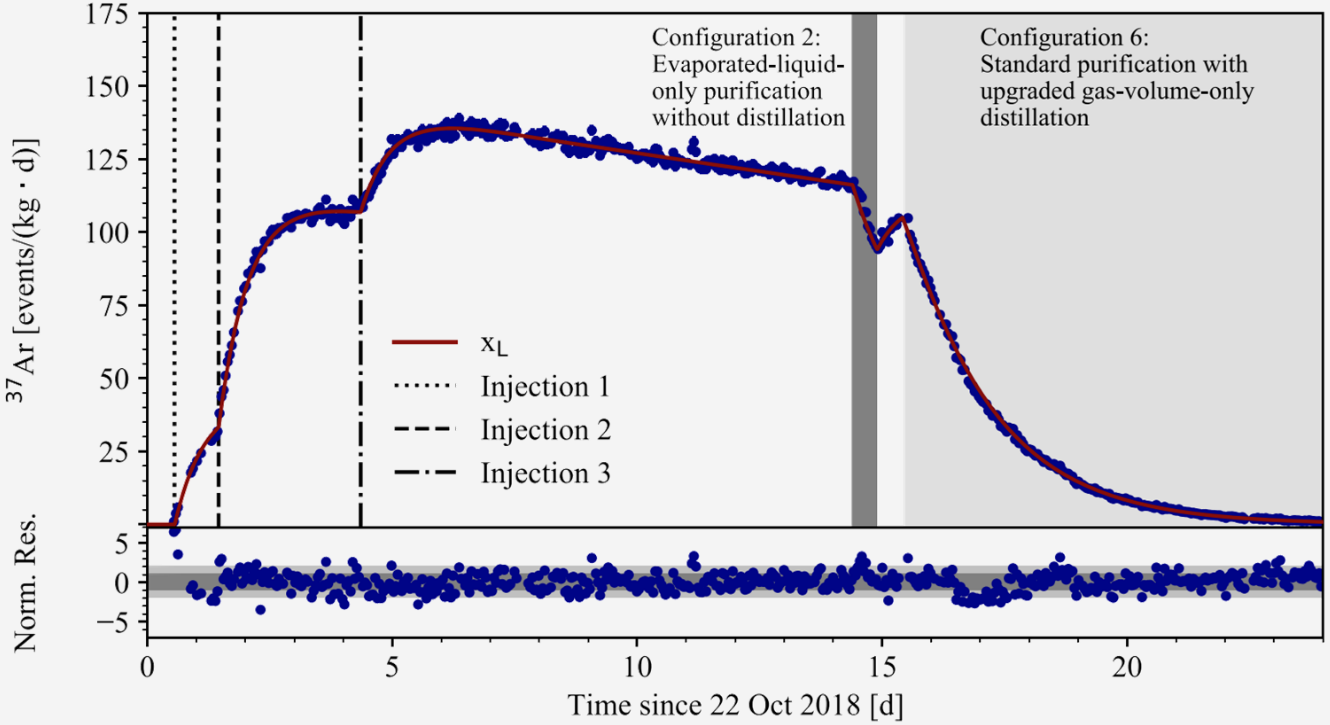
Eur. Phys. J. C (2014) 74:2746

Online Argon removal method for XENON1T

^{37}Ar calibration source:

- A gaseous ^{37}Ar source was deployed in XENON1T in October 2018
  Eur. Phys. J. C 83, 542 (2023)
- ER calibration at 2.8 keV and 0.27 keV (from EC)
- Half-life of 35 days – too long for regular use





- Online Ar distillation in standard Kr removal configuration mode
- Reduction of the ^{37}Ar event rate with effective time constant of $\tau_{\text{eff, Ar}} = 1.7$ d
- Enabled ^{37}Ar as calibration source in XENONnT

Radon removal system for XENONnT

Top Condenser

Custom bath-type LN_2/GXe heat exhanger

Package Tube

Large surface package material

Auxiliary

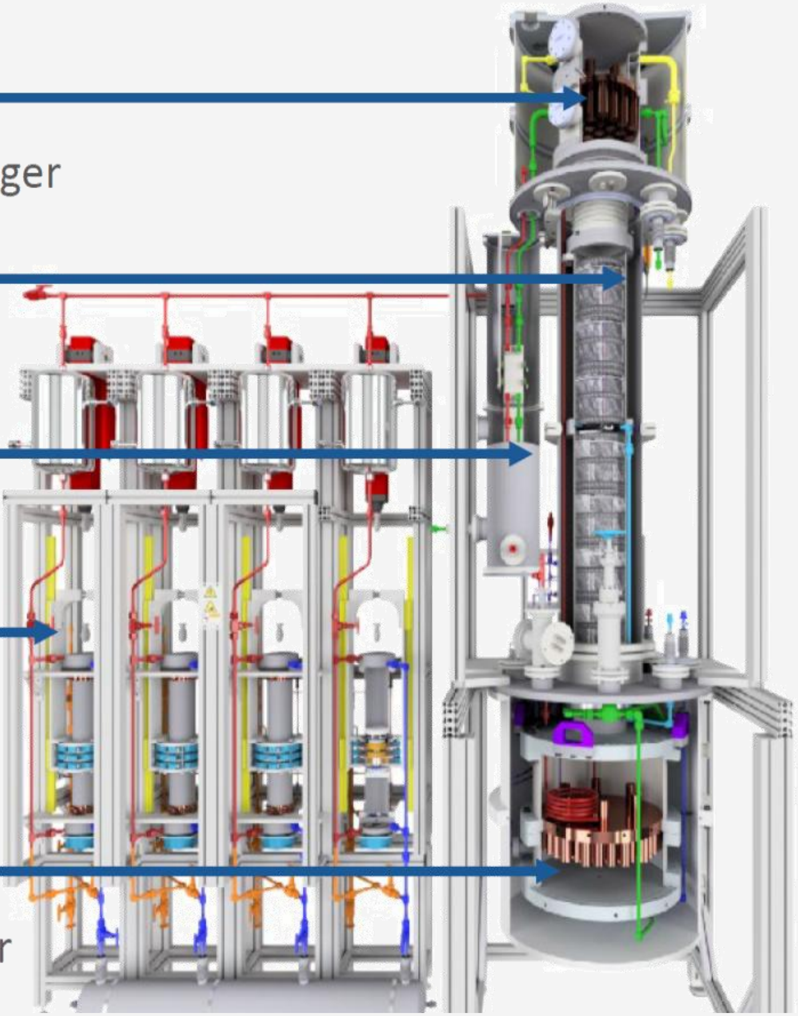
Commercial GXe/GXe heat exchangers

Compressor

Custom four cylinder magnetically-coupled piston pump

Reboiler

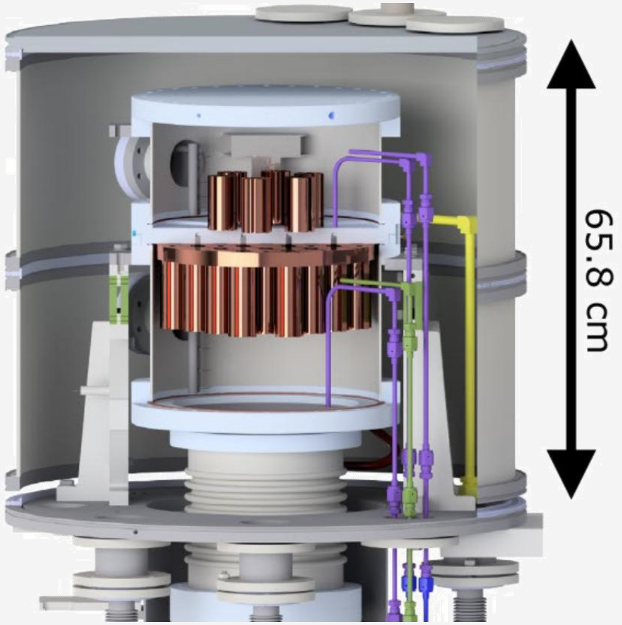
Custom bath-type Xe/Xe heat exhanger

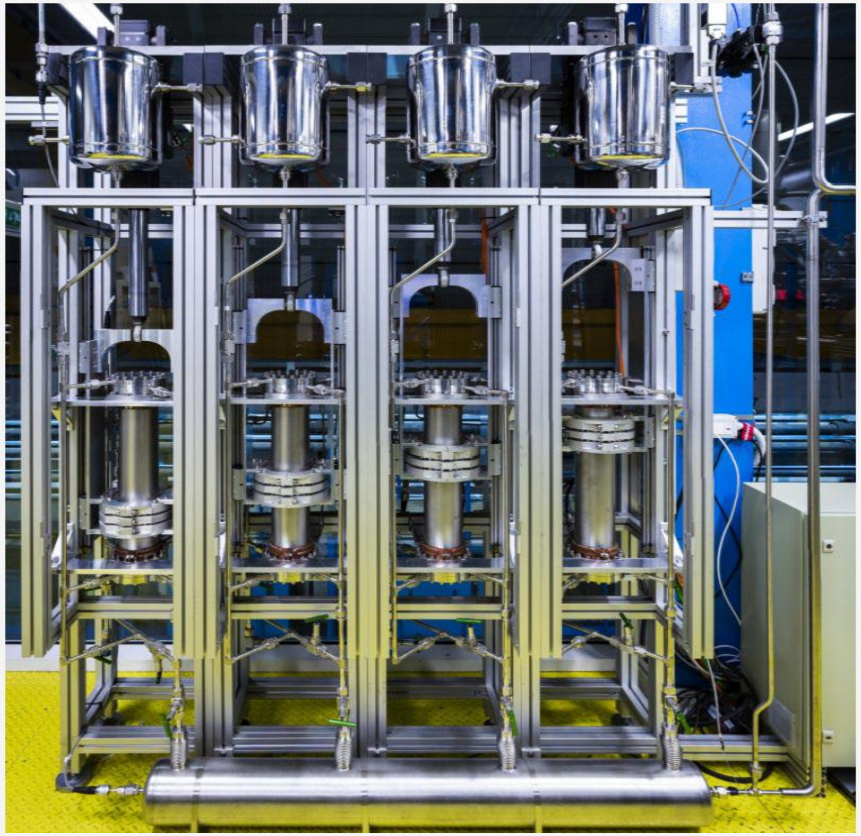


Radon removal system for XENONnT

Top condenser JINST 17 P05037 (2022)

- Custom bath-type LN_2/GXe heat exchanger
- Large surface OFHC copper fins
- **Cooling power:** > 3 kW (required 1 kW)
- **Xe liquefaction rate:** > 113 kg/h (300 slpm) (required 36 kg/h)
- **Cooling efficiency:** 0.98
- **LN_2 consumption:** 660 kg/d @ 1 kW cooling power





Compressor JINST 16 P09011 (2022)

- **Magnetically-coupled piston pump** based on XENON1T prototype: EPJ C78 604 (2018)
- Four piston pumps in **parallel operated phase-shifted**
- **Flow:** 170 kg/h (474 slpm) (required 72 kg/h or 200 slpm)
- **Compression:** 1.8 bar (required 1.5 bar)