

# Krypton Removal from Nitrogen (for Ultra-Low Background Liquid Scintillator Detectors)

## 窒素中のクリプトン除去に関する研究

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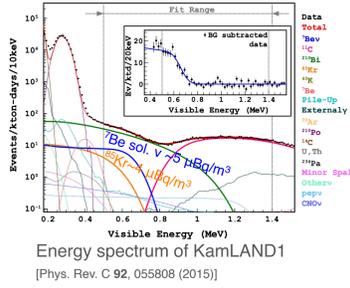
### Key messages

- Commercial LN<sub>2</sub> must be assayed before KamLAND2 operation.
- A TSP makes RGA-based Kr measurement practical by removing bulk N<sub>2</sub>.
- A first CE sample gives a <sup>84</sup>Kr concentration of ~0.94 ppb (preliminary).
- The current sensitivity is ~200 ppt; ~1 ppt is the next realistic target.
- The next milestone is cryogenic adsorption and breakthrough measurement.

## 1. Motivation and required Kr level

### Physics requirement

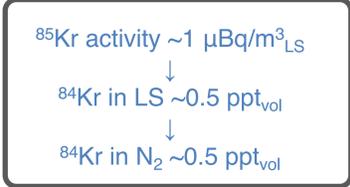
- In KamLAND1, a dedicated N<sub>2</sub> generator system achieved Kr < 1ppt and supported the <sup>7</sup>Be solar neutrino measurement.
- N<sub>2</sub> is used for Liquid scintillator (LS) purging, so Kr in N<sub>2</sub> propagates into LS.
- For KamLAND2, <sup>85</sup>Kr background should remain at or below the 1 μBq/m<sup>3</sup> scale.



Energy spectrum of KamLAND1 [Phys. Rev. C 92, 055808 (2015)]

### Operational context

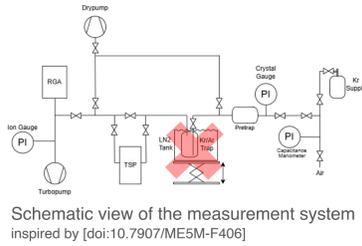
- The original N<sub>2</sub> generator is no longer available.
- KamLAND2 operation relies on commercial LN<sub>2</sub> delivered to a cold evaporator (CE) tank.
- The Kr concentration in CE-N<sub>2</sub> has not been directly measured.
  - KamLAND1 data from a short period of operation with CE-N<sub>2</sub> indicate a <sup>84</sup>Kr concentration of order 0.5 ppb.



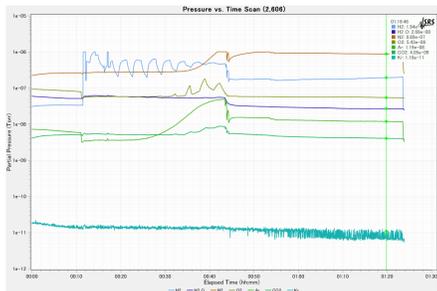
## 2. Measurement strategy: RGA + pre-concentration

### Why pre-concentration is required

- The Residual Gas Analyzer (RGA) is operated at 10<sup>-5</sup> torr and has a dynamic range of ~10<sup>6</sup>.
- Kr is trace even in air, so direct ppt measurement in N<sub>2</sub> is not practical.
- The key idea is to remove bulk N<sub>2</sub> while leaving noble gases for the RGA.



Schematic view of the measurement system inspired by [doi:10.7907/ME5M-F406]



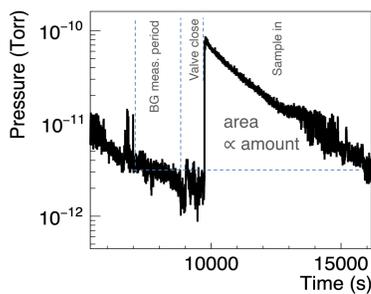
Example of RGA data. Partial pressure of each mass can be measured.

### Cold trap vs TSP

- Cold-trap trials with Molecular Sieve 13X did not provide clean N<sub>2</sub>/Kr separation in the current setup.
- Temperature control was limited to LN<sub>2</sub> on/off, preventing fine desorption control.
- The Titanium Sublimation Pump (TSP) worked: Ti sublimates at ~0.1 g/h, and 0.1 g Ti removes ~20 mL (STP) of N<sub>2</sub> while noble gases are not pumped.



TSP inside a chamber

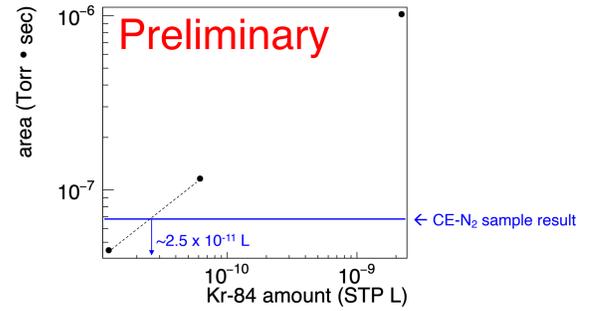


<sup>84</sup>Kr pressure vs time measurement for an air sample (3.4 mL under 2800 Pa)

## 3. Calibration and first CE result

### Calibration quantity

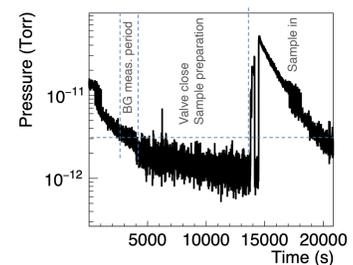
- The integrated <sup>84</sup>Kr signal from the RGA (partial pressure x time, torr s) is mapped to an absolute <sup>84</sup>Kr amount (STP L).
- Three calibration points have been obtained using ambient air and diluted-air samples at fixed injected volume.
  - <sup>84</sup>Kr concentration in air 0.65 ppm
- Uncertainty evaluation is ongoing. Sources:
  - Precision of sampling amount
  - Background fluctuation



RGA signal integration vs absolute <sup>84</sup>Kr amount. Black points are calibration points obtained using air. Blue line is a RGA measurement result for a CE-N<sub>2</sub> sample.

### Sampling and measurement of CE-N<sub>2</sub>

- CE-N<sub>2</sub> (~0.4 MPaG) was collected into a 150 mL cylinder.
- The cylinder was purged with 10 L of CE-N<sub>2</sub> before sampling.
- 26.7 mL (STP) from the cylinder was processed by the TSP-RGA system.



<sup>84</sup>Kr pressure vs time measurement for CE-N<sub>2</sub> sample (26.7 mL STP)

### Preliminary CE-N<sub>2</sub> result

- ~2.5 x 10<sup>-11</sup> L of <sup>84</sup>Kr in 26.7 mL of CE-N<sub>2</sub>
- <sup>84</sup>Kr concentration 0.94 ppb
- Consistent, at the order-of magnitude level, with the ~0.5 ppb indication from a short CE-N<sub>2</sub> period data in KamLAND1.
- CE-N<sub>2</sub> likely requires additional purification to reach the ppt goal.

## 4. Sensitivity and path to the 1 ppt goal

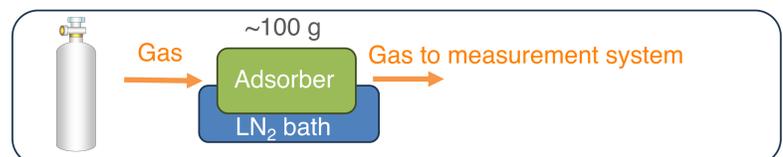
- Sensitivity is set by the RGA background and by the processed sample amount.
- Current conditions:
  - Background ~2 x 10<sup>-12</sup> torr
  - Sample amount ~30 mL (STP)
  - giving a concentration sensitivity of ~200 ppt.
- Expected improvements:
  - Careful bakeout may lower the background to ~1 x 10<sup>-13</sup> torr
  - Fresh TSP filaments should allow at least ~300 mL (STP) to be processed.
- Together these upgrades → ~1 ppt sensitivity

## 5. Next step: adsorption test and implementation concept

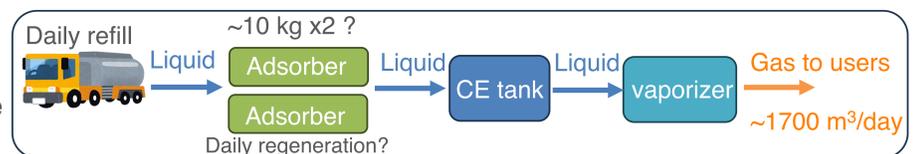
### Cryogenic adsorber study

- Candidate adsorbents: activated carbon and silver-zeolite
- In laboratory tests, GN<sub>2</sub> will be passed through adsorbents cooled to LN<sub>2</sub> temperature.
- In KamLAND2 process, LN<sub>2</sub> will be passed through adsorbents.
- It may be necessary to study difference between GN<sub>2</sub> and LN<sub>2</sub> purification.
  - Decrease of adsorption capacity due to liquefaction of N<sub>2</sub> is indicated in [1].

Laboratory Test



Planned process in KamLAND site



[1]: G. Zuzel, "Searching for rare nuclear processes at low energies – development of background reduction techniques for cutting-edge Experiments" (2017)