



Low background R&D for DARWIN

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DARWIN background specifications

- Goal is to be completely neutrino dominated: JCAP11 (2016) 017
- Neutron-induced nuclear recoil (NR) background negligible
 - But requires radiopure materials
- Coherent solar neutrino scattering will be irreducible NR background
- Also Electronic recoil (ER) background dominated by solar neutrinos
- All other components to be at ~10% level
 - Most critical are dissolved impurities, in particular ²²²Rn (Factor 10 reduction w.r.t. XENONnT)



⁸⁵Kr control for DARWIN

- Xenon is produced from air → Traces of krypton (~50 ppb)
- ⁸⁵Kr due to anthropogenic nuclear activities: 85 Kr/ nat Kr = 2x10⁻¹¹
- Goal for Kr in Xe 30 ppq (1ppq = 1x10⁻¹⁵)
- Requires powerful Xe purification technique (distillation) and extremely sensitive analytics







Kr removal in XENON1T / XENONnT

- Kr column built for XENON1T
- Measured separation ~640.000 with 99% Xe recovery: EPJC (2017) 77:275
- Stable up to 6.5 kg/h throughput
- Lowest achieved concentration: 26 ppq
- Outlet concentration sufficient for DARWIN
- Sufficient for online distillation, but larger throughput needed for initial Xe purification
- Reliable Kr/Xe analytics with ppq-sensitivity required



Sep 15, 2022



Rare Gas Mass Spectrometry (RGMS) at MPIK

- Requires efficient removal of Xe \rightarrow gas chromatography.
 - Homemade UHV-qualified system
 - Low mass packed columns
 - Thoroughly purified carrier gas (helium)
- ► → 8 ppq detection limit: EPJC (2014) 74:2746
- Future: Automated RGMS (Auto-RGMS)
 - Fully automated operation
 - Optimized separation columns
 - Improved repeatability and robustness
 - Design is finished, construction has started
 - Expect commissioning in summer 2023
 - Eventually on-site online operation?





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Radon mitigation techniques



- ²²²Rn is produced in situ by emanation from detector materials
- ²²²Rn mitigation by:
 - Avoiding:
 - Material screening and selection
 - Removal of ²²²Rn sources
 - Purification:
 - Distillation / Adsorption
 - Tagging
 - Analysis techniques

Material screening and selection



- Standard technique is HPGe gamma spectroscopy
- World's most sensitive spectrometers are owned by DARWIN groups: GeMPI I, II, II, IV, GATOR, GeMSE, GIOVE, ...
- ²²⁶Ra/²²⁸Th detection limit down to 10 µBq/kg
- But ²²²Rn emanation rate is NOT strongly correlated to ²²⁶Ra bulk contamination
- → Dedicated ²²²Rn screening facilities

Material screening by ²²²Rn emanation assay



- ²²²Rn-emanation rate provides complementary information
- ²²²Rn screening in DARWIN:
 - Electrostatic radon monitors
 - Ultralow background miniaturized proportional counters
 - Sensitivity: ~10 ²²²Rn atoms
- @MPIK: Fully automated ²²²Rn concentration system (AutoEma) from up to 80 liters emanation chambers





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Ongoing R&D related to radon emanation



- Better understanding of emanation process.
- Recoil-driven versus diffusion-driven emanation
 - Dependency on temperature
 - Dependency on envrionment
 - Gas (helium, xenon, air, vacuum)
 - Humidity
 - Gaseous versus liquid
- Measurement of radon diffusion constant in different materials
 - Diffusion constants <10⁻²² cm²/s in metals
- Radon self-trapping in porous materials

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Understanding ²²²Rn source distributions

Example: XENON1T setup



Understanding ²²²Rn source distributions

Example: XENON1T setup



Understanding ²²²Rn source distributions

Example: XENON1T setup



Hermetic TPC with quartz chamber

- Hermetic TPC avoids ²²²Rn contamination from outer volume
- Also reduces electro-negaative impurities
- Quartz is suitable chamber material
 - Low intrinsic radioactivity
 - UV-transparent
 - Avoids secondary electron emission
- Succesful operation of prototype as dual-phase TPC
- Ongoing: Optimization of hermeticity (quartz flange sealing)

Hermetic TPC with more conventional design

- Modified standard TPC design
- Sealing by cryofitting: Large PTFE shrinkage at low temperature seals against metals
- Succesful dual-phase TPC operation: Good performance
- Hermeticity test with ^{83m}Kr:
 - ~0.1 kg/h leakage rate
- Further improvement on sealing possible

arXiv: 2209.00362

Scaling up: What does it mean for DARWIN?

arXiv: 2209.00362

- Upscaling of results for 10⁵ times larger DARWIN TPC
- Location of leak crucial
- Assumptions on ²²²Rn emanation rates based on XENON1T / XENONnT screening results
- Online Rn purification included in model
- Darwin goal for ²²²Rn is reachable, but requires combination of all ²²²Rn reduction techniques

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Radon removal by xenon distillation

- Radon is less volatile than xenon -> reverse operation mode w.r.t. Kr removal
- Feasibility demonstrated in Xenon100: EPJC (2017) 77:358
- Applied in XENON1T: 4.5 μBq/kg achieved: EPJC (2021)81:337
- High flow radon removal system developed for XENONnT: arXiv: 2205.11492
- 1.7 µBq/kg achieved

- Upscaling only possible for high throughput system
- Processing speed for DARWIN must be >=10 tons/day
- Efficiency in power consumption and xenon holdup versus radon reduction is crucial

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Rn-free Xe out

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Radon mitigation by surface coating

- A thin ²²²Rn-tight, clean (²²⁶Ra-free) surface coating should block ²²²Rn
- Should work for recoil-driven AND diffusion-driven emanation
- Various techniques investigated: Vapor deposition (PVD, CVD), plasma etching, electrochemical coating

Electrochemical copper coating

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- Initial tests with ²²⁰Rn (thoriated welding rods and recoil-implanted stainless steel samples)
- Procedure for stable Cu coating was developed inhouse at MPIK over the last few years
- Optimization (surface current / adhesion layer, ...)

Coating of ²²²Rnemanating SS disc

- ²²⁶Ra implanted SS disc produced at ISOLDE facility (CERN)
- 30 keV implantation energy
 - ► \rightarrow 8 nm mean depth
- Full characterization of sample (α and γ -spectroscopy, ²²²Rn emanation rate)
- arXiv: 2205.15926
- ²²²Rn emanation rate:
 - Before coating: (2.00 ± 0.05) Bq
 - After coating: (4.3 ± 0.3) mBq
- ²²²Rn reduction factor: ~470

Coated ISOLDE sample: Temporal development of ²²²Rn emanation rate

- ²²²Rn emanation rate was found to decrease
- Coating is getting "tighter"
- Oxidation?
 - But storage under protective atmosphere
- Re-crystallisation?
- Final ²²²Rn reduction factor: ~1700
- Next step: Upscaling

Summary

- Ultimate DARWIN experiment require ultiamtely low background level
- Dissolved noble gases dominate radioactive background
- Kr-control by cryo-distillation and ultrasensitive analytics techniques
- ²²²Rn control is most challenging.
- 0.1 µBq/kg will can only be achieved by combining
 - careful material selection
 - powerful online purification
 - novel ²²²Rn mitigation techniques

Comparison of sensitivity (following procedure outlined in EPJC (2015) 75:531)

					sample chamber configuration								
		class 1: small low-mass samples represented by point-source				class 2:	large hig copper ble	h-density ock	samples	represent	ed by		
			threshold activity [mBq]					threshold activity $[mBq/kg]$					
/-	Isotope	Bruno	Corrado	GIOVE	GeMSE	Gator	GeMPI II	Bruno	Corrado	GIOVE	GeMSE	Gator	GeMPI II
	²³⁸ U	56.08	39.95	12.01	3.64	2.24	1.86	168.57	16.26	3.23	1.22	0.68	0.67
	²²⁶ Ra	1.06	0.70	0.19	0.069	0.083	0.042	3.75	0.36	0.071	0.030	0.029	0.019
/	²²⁸ Th	1.69	1.15	0.33	0.12	0.13	0.081	3.99	0.37	0.046	0.031	0.027	0.020
	228 Ra	1.74	1.19	0.35	0.11	0.11	0.084	5.32	0.49	0.11	0.037	0.032	0.030
	$^{137}\mathrm{Cs}$	0.59	0.44	0.12	0.026	0.040	0.027	2.16	0.22	0.034	0.011	0.015	0.012
	$^{60}\mathrm{Co}$	0.40	0.28	0.13	0.066	0.028	0.023	1.03	0.09	0.025	0.016	0.0064	0.0062
	40 K	11.60	4.07	1.68	0.47	0.50	0.35	29.96	1.34	0.22	0.13	0.12	0.10

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Optimization of Cu coating procedure and ²²⁰Rn reduction results

- Optimum surface current density identified
- Avoid whisker growing by careful parameter control

- ²²⁰Rn reduction factor up to 100 achieved for 5 μm thick Cu coating
- Hints for ²²²Rn reduction
- Recoil emanation fully stopped
- Diffusion-driven emanation confirmed by tests at different temperatures

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Coating (and de-coating) of ²²⁴Ra-implanted SS disc

- ²²⁴Ra decays on SS disc
- ²²⁰Rn is emanated and decays in electrostatic radon monitor
- Charged ²¹²Pb (²²⁰Rn-daughter) is collected on PIN diode
- Counting of ²¹²Po alpha-decays
- ²²⁰Rn reduction factor R: 20<R<1000
- Upper limit from coating, lower limit from de-coating