Low Radioactivity Techniques (LRT 2024)

Kraków, Poland, 01-04 October 2024

Purification strategy of the JUNO liquid scintillator

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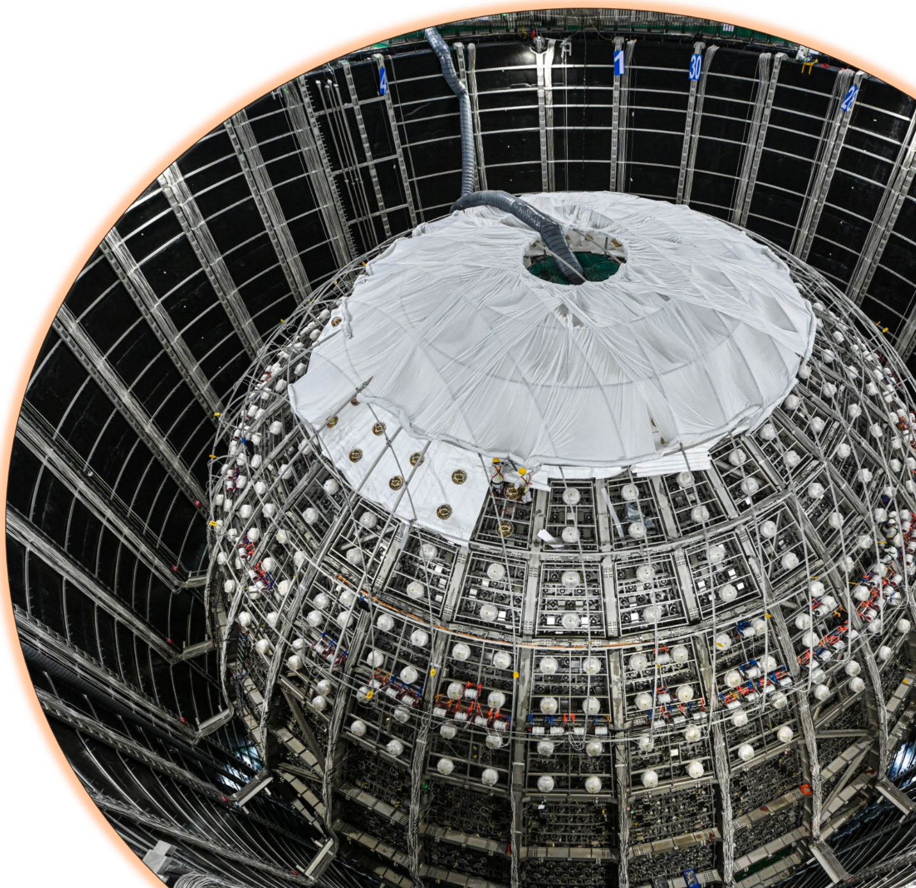
on behalf of the JUNO Collaboration

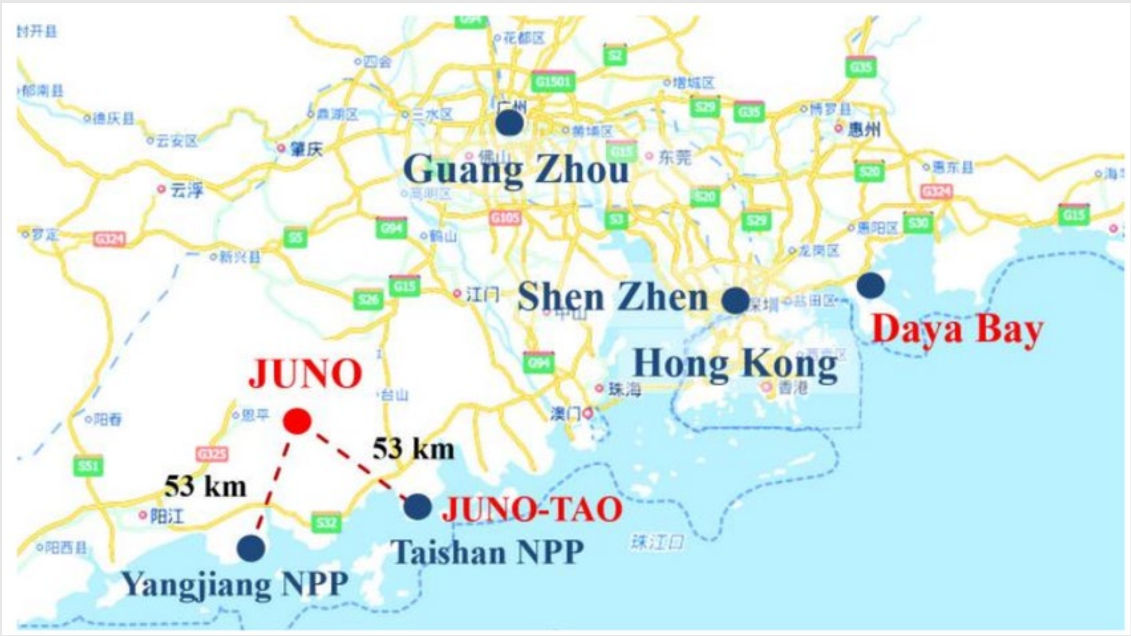


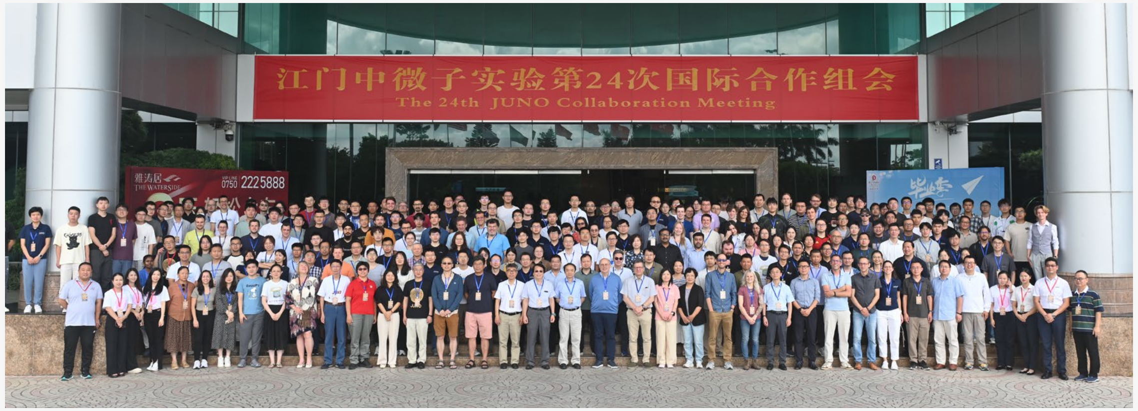
Jiangmen Underground Neutrino Observatory

JUNO is a **20 ktonnes** multi-purpose underground liquid scintillator detector

- Currently under construction in Southern China
- **Main goal**: determine the NMO and ν oscillation parameters, detecting ν from several sources

[JUNO physics and detector, PPNP, 123 (2022) 103927]





Huge collaboration: >700 collaborators, 74 institutes in 17 countries/regions

JUNO detector

Central detector (CD):

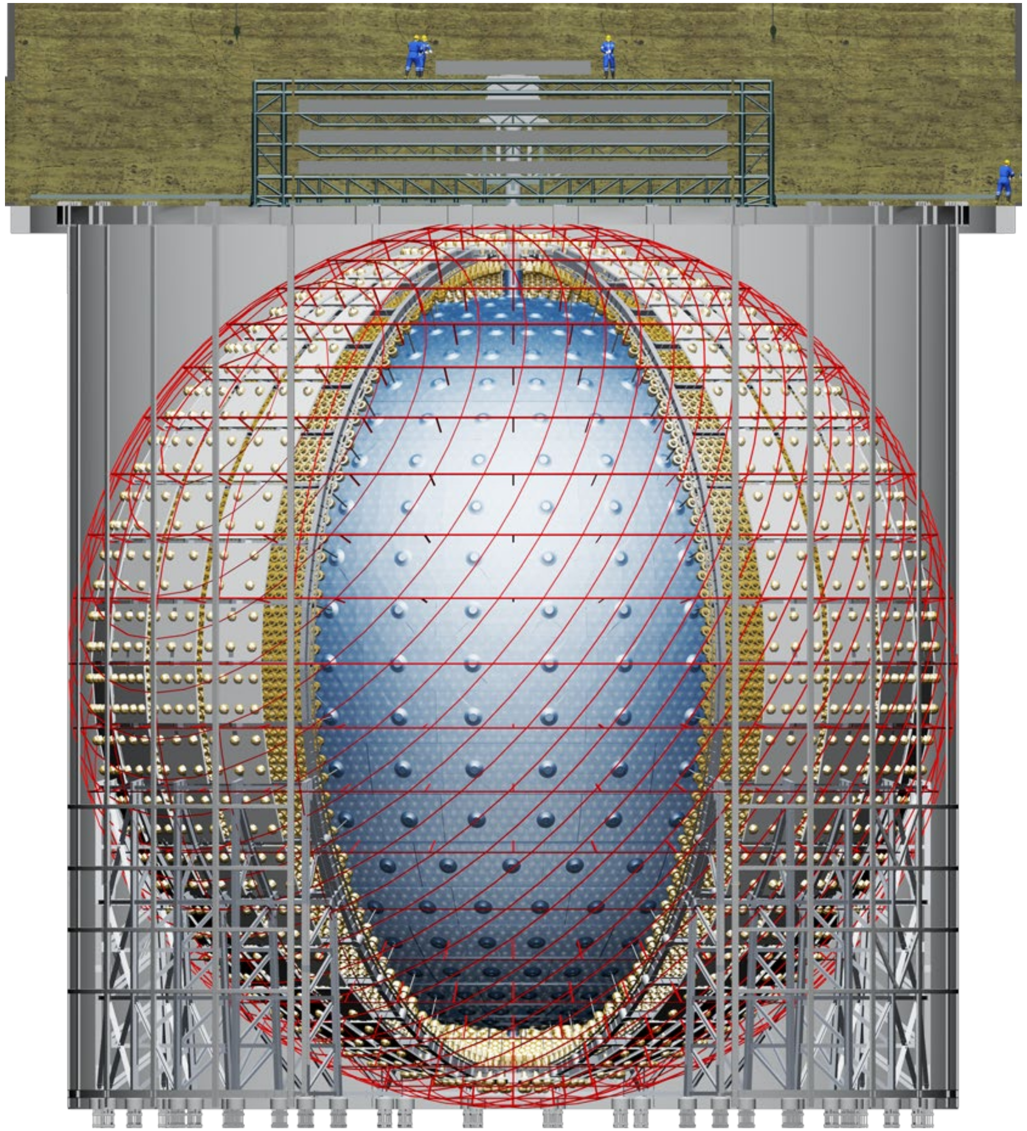
- 35,4 m-diameter acrylic sphere (thickness: 12 cm)
- 20.000 tonnes liquid scintillator (LS) as target
- 17.612 large PMTs (20-inch)
- 25.600 small PMTs (3-inch)
- 78% coverage

Water pool (WP):

- Muon veto + shield against external radioactivity
- 35-kton ultra-pure water
- 2.400 large PMTs (20-inch)

Top tracker muon veto:

- 3 layers of plastic scintillator



Detector technology and needs

Which are the key features and challenges for JUNO?

- **Huge active mass** (largest ever LS detector)
- **Excellent light propagation and collection**:
  - LS optical properties
  - PMTs (78% coverage)
- **Low background experiment**:
  - veto and shielding against external radioactivity
  - material selection
  - underground facility
  - reduction of LS internal radioisotopes

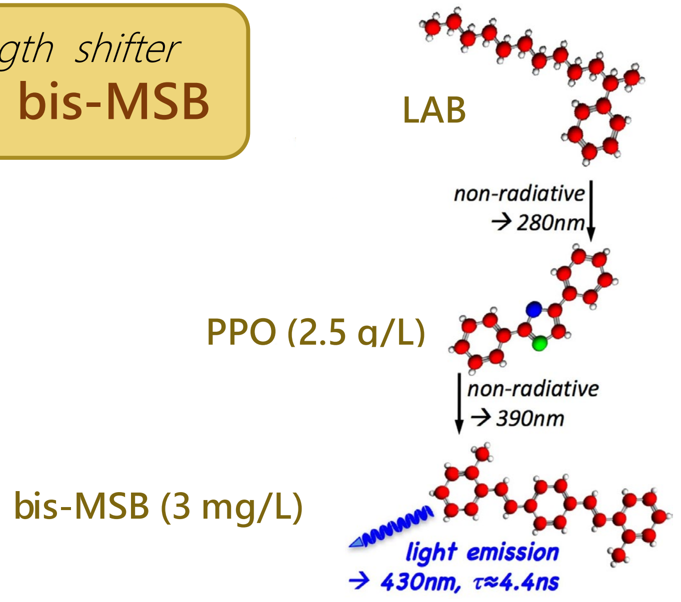
These can be significantly improved by purifying the scintillator with dedicated purification systems

⇓

Unprecedented energy resolution: 2.95% @1 MeV

JUNO LS requirements

LS recipe : *solvent* Linear Alkyl Benzene (LAB) + *fluor* 2,5 g/L PPO + *wavelength shifter* 3 mg/L bis-MSB

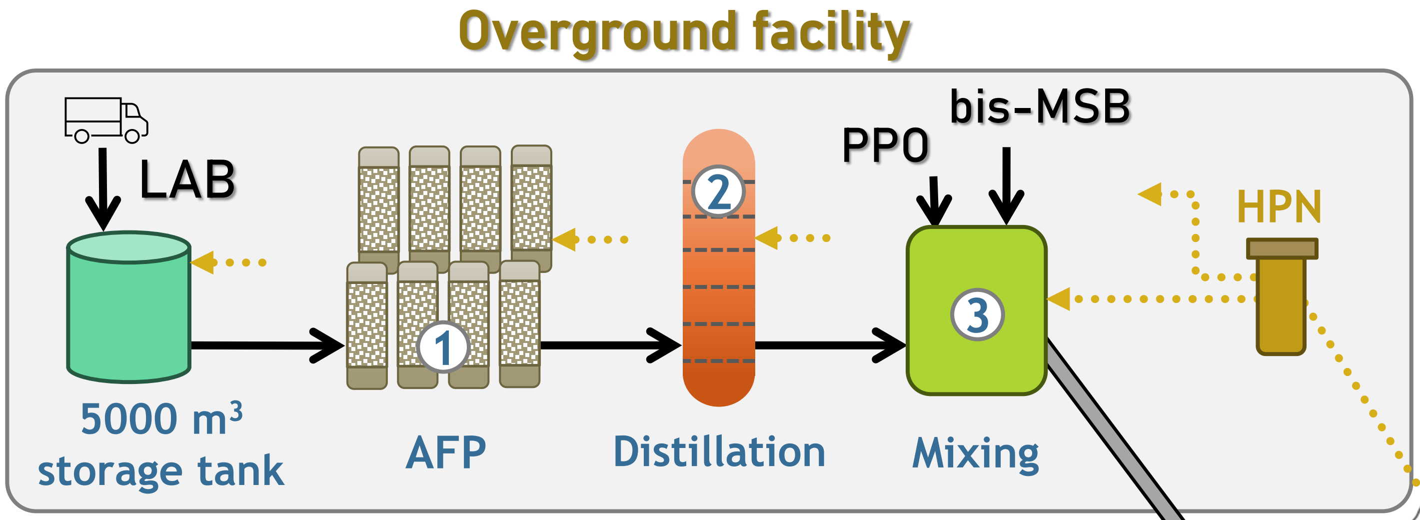


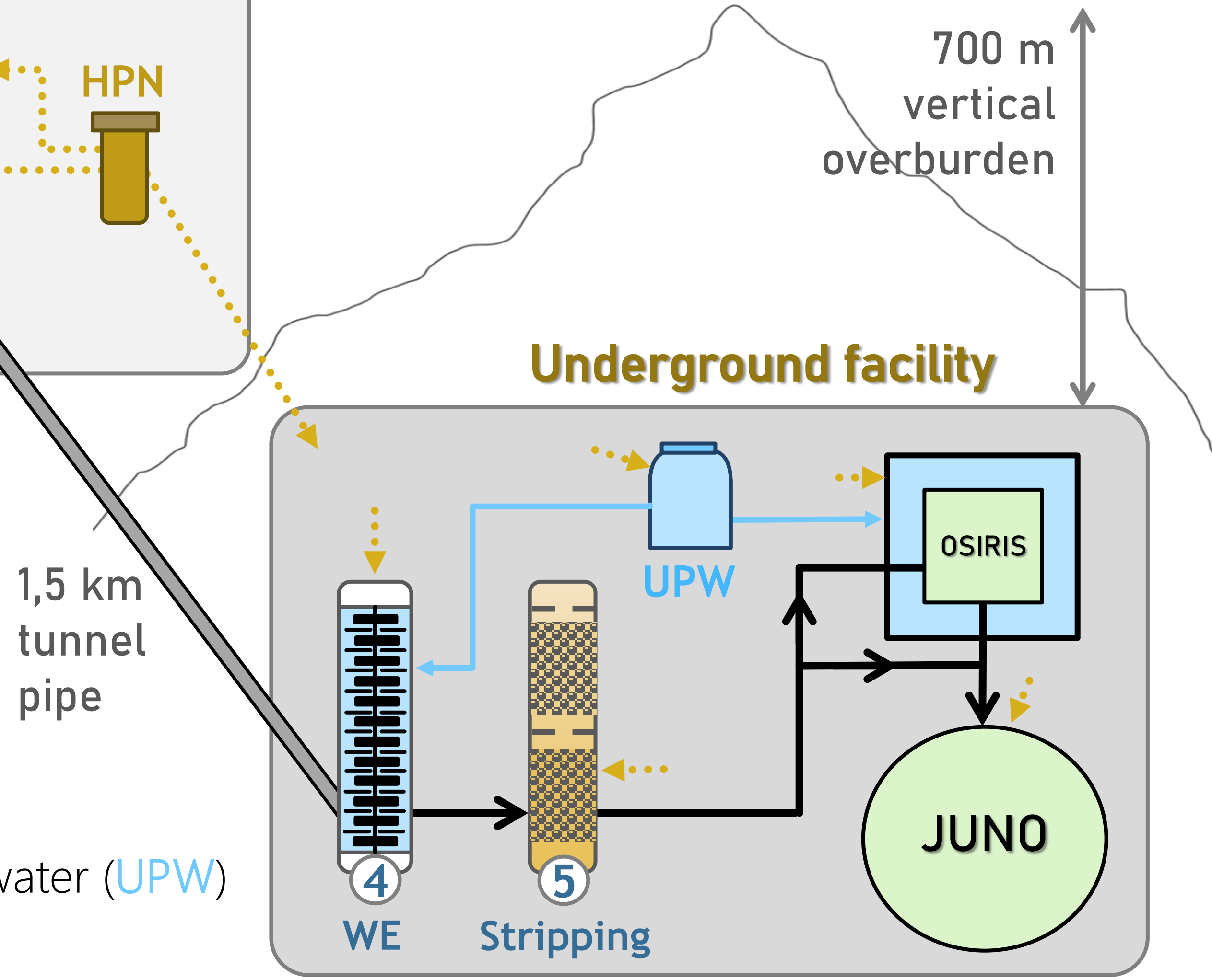
- Optical requirements:
  - High light yield: ~10.000 Photons/MeV → ~1665 p.e./MeV
  - Attenuation length: LAB > 23 m; LS > 20 m @430 nm
- Radio-purity requirements:
  - Reactor anti-neutrinos: $^{238}U/^{232}Th < 10^{-15}$ g/g
  - Solar neutrinos: $^{238}U/^{232}Th < 10^{-17}$ g/g
- Other requirements:
  - Energy response linearity
  - Long term stability (ageing)

See Xin Ling's poster

Radioisotope	Contamination source	JUNO requirement	
		minimum	ideal
^{222}Rn	Air and emanation from material	$< 250\,\mathrm{mBq/m^3}$	$< 5\,\mathrm{mBq/m^3}$
^{238}U	Dust suspended in liquid	$< 10^{-15}$ g/g	$< 10^{-17}$ g/g
^{232}Th	Dust suspended in liquid	$< 10^{-15}$ g/g	$< 10^{-17}$ g/g
^{40}K	Dust suspended in liquid, PPO	$< 10^{-16}$ g/g	$< 10^{-18}$ g/g
^{39}Ar	Air	$< 50\,\mu\mathrm{Bq/m^3}$	$< 50\,\mu\mathrm{Bq/m^3}$
^{85}Kr	Air	$< 4\cdot10^{-24}$ g/g	$< 8\cdot10^{-26}$ g/g

JUNO purification procedure



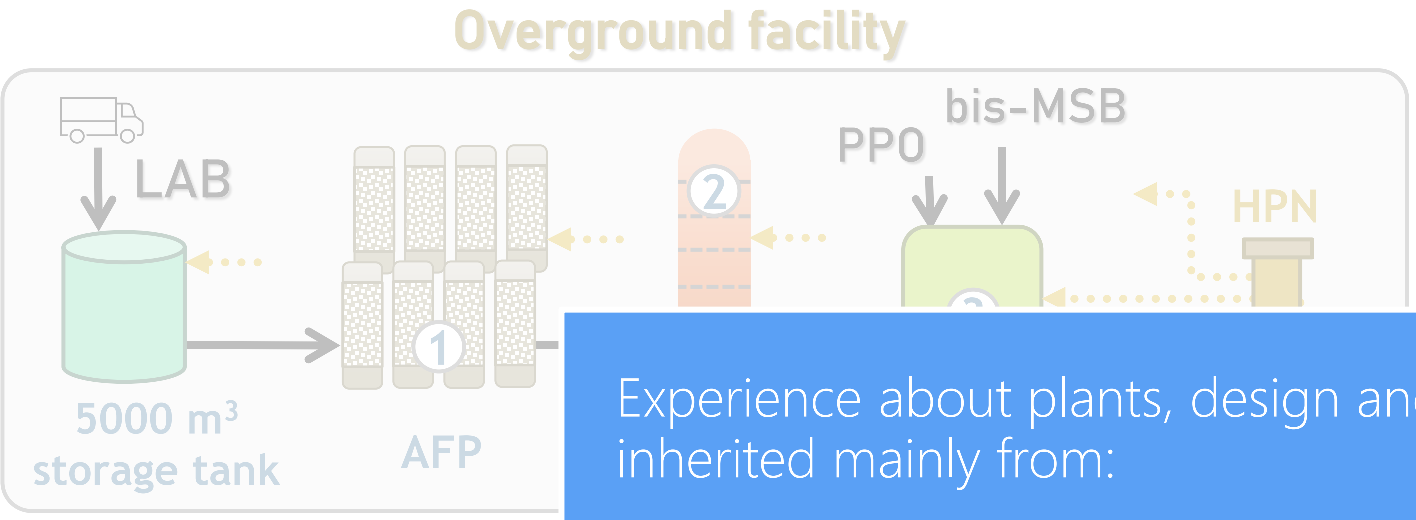


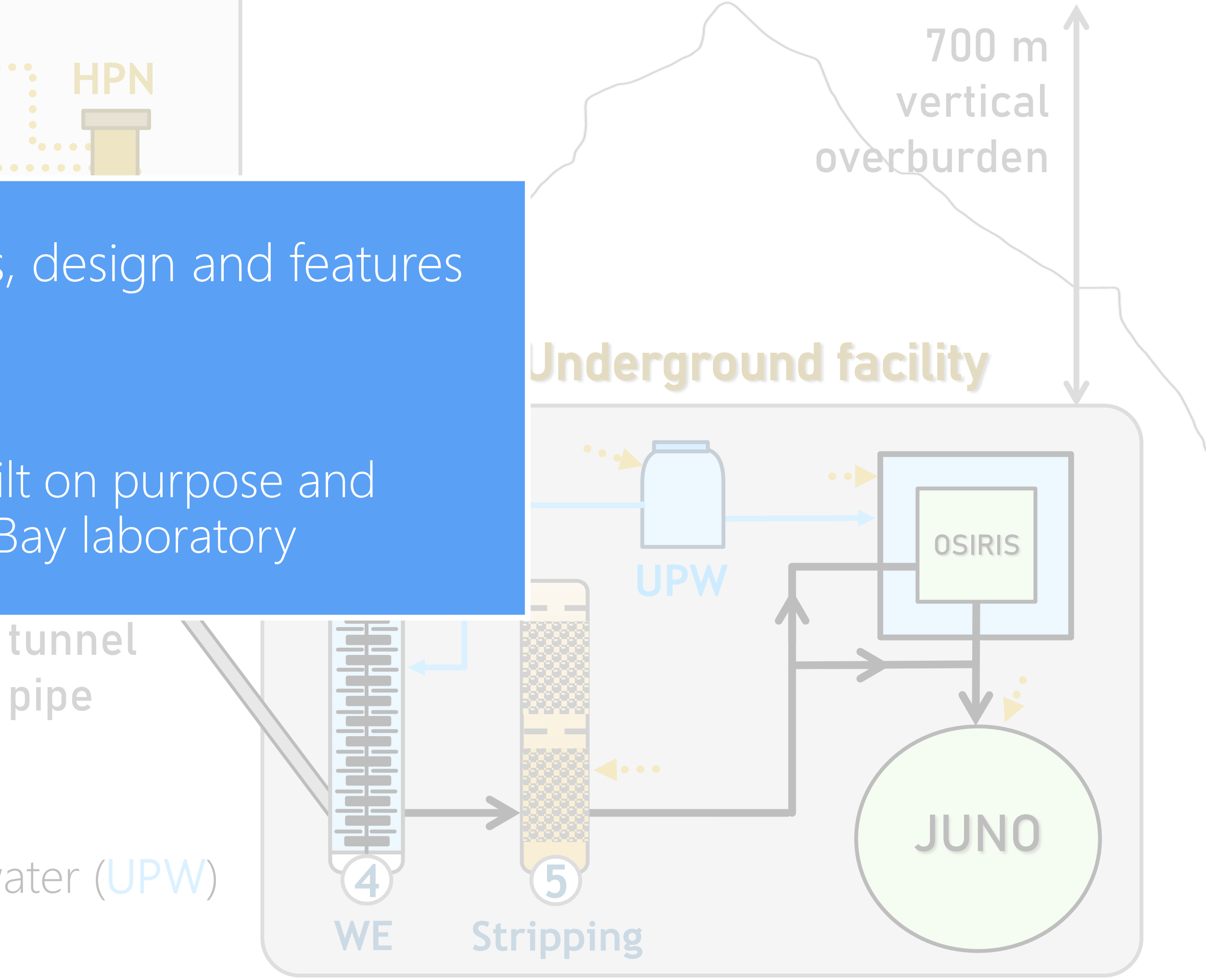
5 purification large-scale plants (7 m^3/h):

1) Alumina filtration plant (AFP)
2) Distillation plant
3) Mixing plant
4) Water Extraction plant (WE)
5) Gas Stripping plant

- Auxiliary systems: high-purity N_2 (HPN), ultra-pure water (UPW)
- Online quality monitoring system: OSIRIS

JUNO purification procedure





Experience about plants, design and features inherited mainly from:

- Borexino
- Small-scale plants built on purpose and installed at the Daya Bay laboratory

5 purification large-scal

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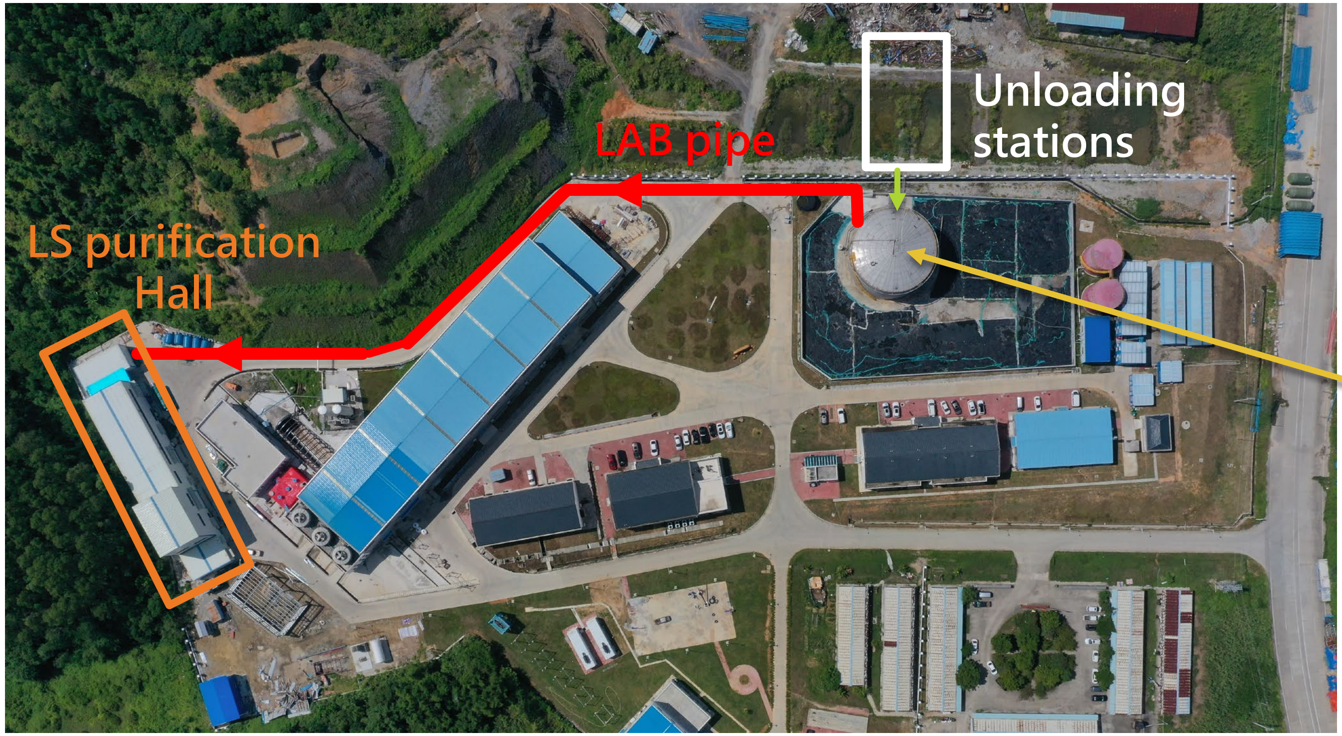
LAB/LS purification plants

LAB transportation & storage

- **High quality LAB**, with special production process by Jinling Petrochemical Co. Ltd
- Transportation to JUNO site via **n.200 ISO tanks** (20 tonnes LAB/each)
- Storage in the big tank:
  - Tank Volume: 5000m³, ~4200 tonnes raw LAB
  - 304L stainless steel with 0.4 um roughness



① Alumina filtration plant (APF)

Goal: - removes optical impurities
- increases the attenuation length of LAB

Overground LS building

⇒ Optical purification of LAB

Filtration through **alumina (Al_2O_3) powder**: removes optical contaminants (oxidized molecules, fused ring compounds)

Operation:

- LAB pumped through **8 columns** containing Al_2O_3 (bed volume BV=0,5m^3/each)
- 7 columns running + 1 always in maintenance
- Powder is replaced with new one, before saturation with removed contaminants
- Double filtration (220 nm, 50 nm)

AFP parameters	
N° columns	8 (7+1)
Height	2,8 m
Diameter	0,6 m
Bed volume (BV)	0,5 m^3
Total flow rate	7 m^3/h
Flow rate per col.	1 m^3/h (2 BV/h)
Filters	220 nm/50 nm

① Alumina filtration plant (APF) - results

Al_2O_3 powder features:

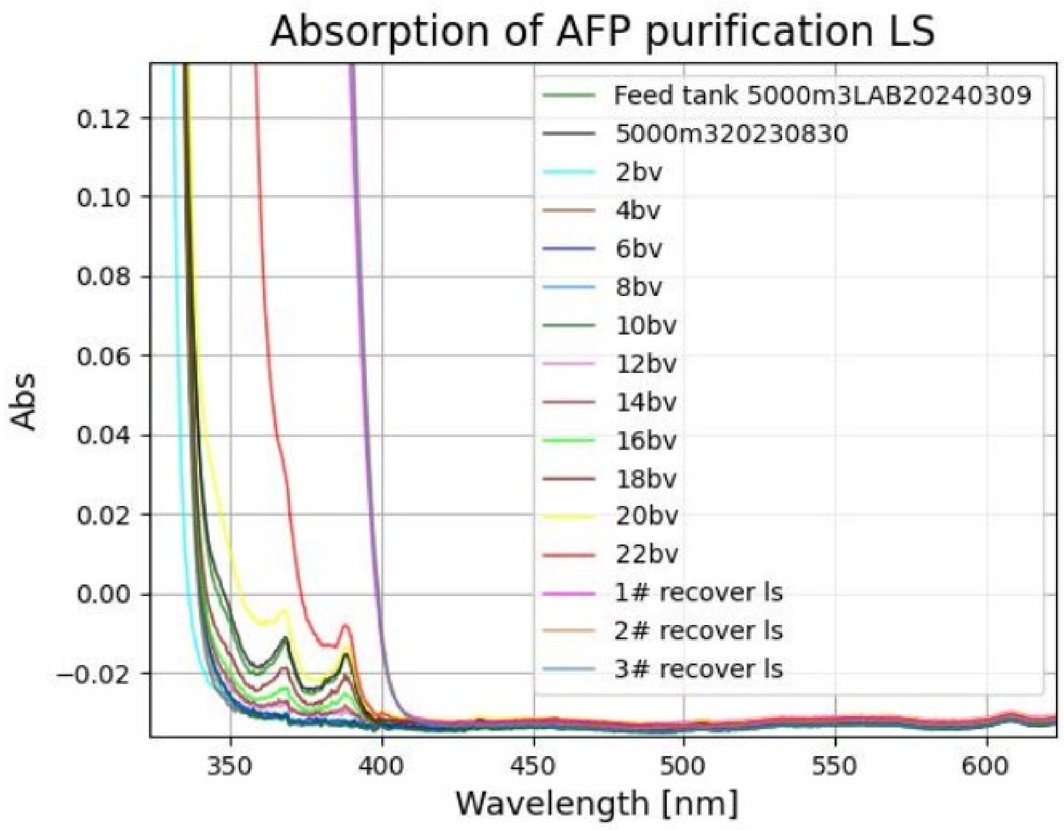
- Double vacuum packaging for transportation
- 20 of 500 tons onsite
- Low radioactivity: < 0.4 Bq/kg in ^{238}U, ^{232}Th

Plant fully commissioned onsite!

Results:

- **Absorption spectra**: successful removal of peaks in 355-400 nm interval (up to 20 BV)
- **Attenuation length**: raw LAB ~20 m → after AFP > 23,5 m
- ^{238}U<0,31 ppq; ^{232}Th=0,9± 0,1 ppq (preliminary results)

[Zhu et al., NIM A, 1048 (2023) 167890]



ppq (parts per quadrillion) = 10^{-15} g/g

② Distillation plant

Goal: - removes heavy metals, ^{238}U, ^{232}Th, ^{40}K
- further improves optical properties of LAB

Overground LS building

⇒ Radiochemical+optical purification of LAB

7m-high distillation column, with 6 sieve trays and internal reflux

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Overground LS building

⇒ Radiochemical+optical purification of LAB

7m-high distillation column, with 6 sieve trays and internal reflux

Column operation:

- Fractional distillation **under vacuum: 5 mbar, 210-220°C**
- Reduced boiling T: no thermal degradation
- LAB boiled in the bottom part. High-boiling, low-volatility impurities accumulated at bottom. Purified vapors extracted at the top

Distillation column parameters	
N° trays	6 (3500 holes/ea)
Height	7 m
Diameter	2000 mm
Temperature	210-220 °C
Pressure	5 mbar
Internal reflux	max 50%
Bottom discharge	max 100 L/h
Thermal power	1 MW_{th}

② Distillation plant - results

Process parameters:

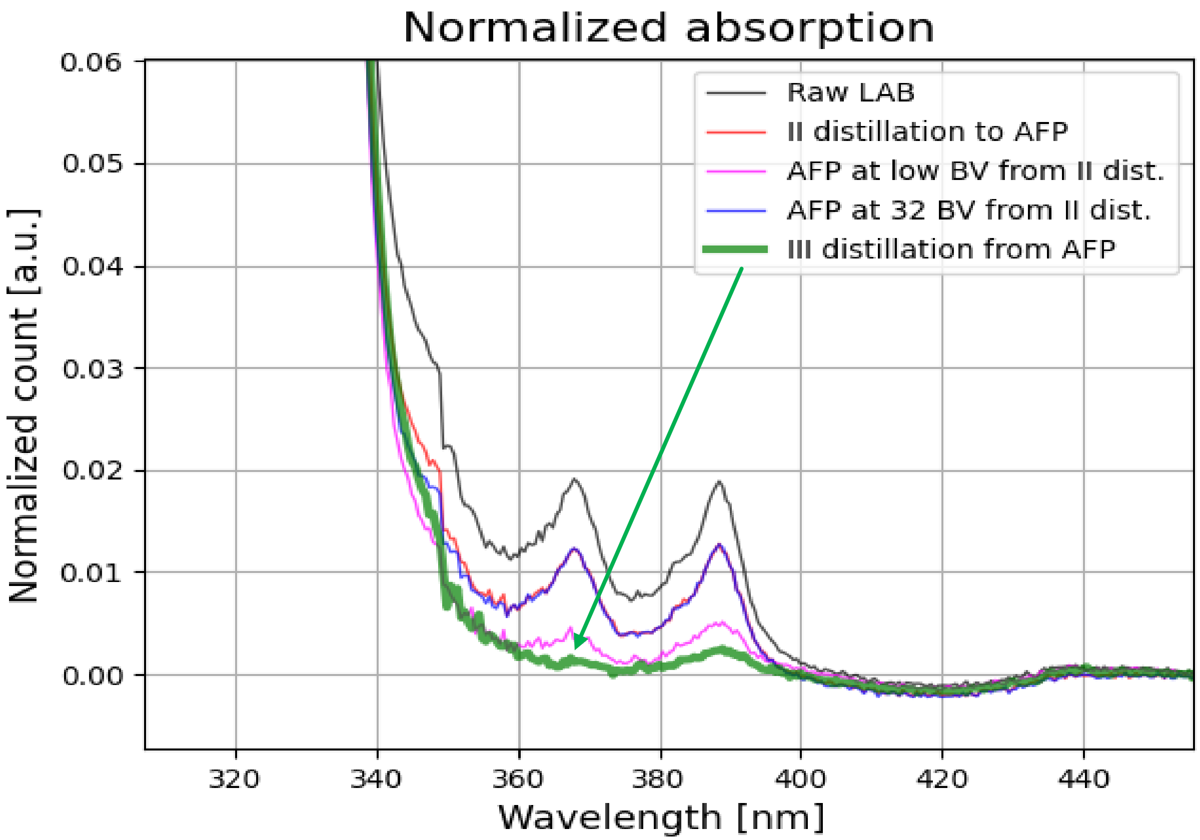
- Top-bottom Δp ~55 mbar
- ~13 cm layer of liquid on each tray (720 kg/m^3 LAB@ 200°C)
- N. theoretical stages: 5

Plant fully commissioned onsite!

Results:

- ^{238}U<0,28 ppq; ^{232}Th<0,22 ppq; ^{40}K<1,7 ppq (preliminary results)
- Absorption peaks in 355-400 nm range further reduced

[Landini et al., NIM A, 10.1016/j.nima.2024.169887]



③ Mixing plant

Goal: - ^{238}U, ^{232}Th removal from PPO and bis-MSB
- production of LS with JUNO recipe

Overground LS building

⇒ Radiochemical purification of PPO, bis-MSB (LS)

? acid extraction + filtration

Plant operation:

- PPO and bis-MSB dissolved in LAB in higher concentration (=Master Solution, **MS: x42 JUNO recipe**)
- **MS purification**: 1 acid extraction with 5%HNO_3 + 2 water rinsing cycles
- **MS double filtration**: functional group filters + 50 nm filters
- Online dilution to JUNO recipe with LAB

Mixing plant parameters	
Master Solution	105 g/L PPO 126 mg/L bis-MSB
Dissolving T	40°C
Acid washing	40°C with 5% HNO_3
N° acid washing	1 time, 1:2 ($2m^3$ acid : $4m^3$ MS)
N° water washing	2 times, 1:1

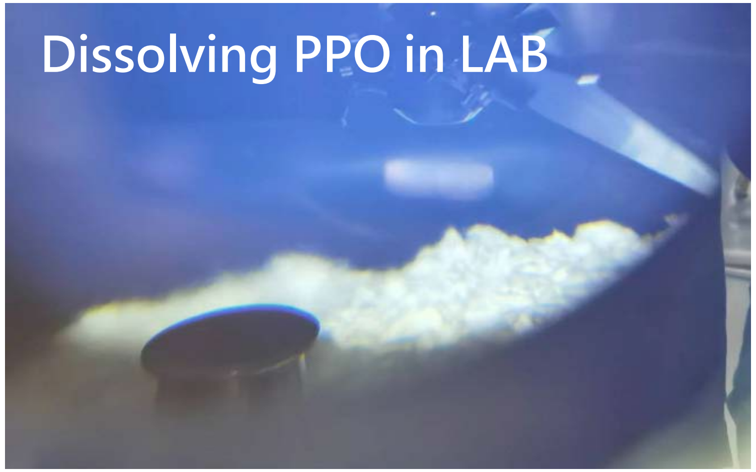
3 Mixing plant - results

[X. Sun., Neutrino2024 conf., Zenodo, 10.5281/zenodo.13684977]

Features and status:

- **60 tonnes PPO** (100% arrived): U=0,066 ppt, Th=0,090 ppq (requir.: <0,43 ppt ✓)
- **72 kg bis-MSB** (28% arrived): U=2,0 ppt, Th=4,1 ppq (requir.: <8,3 ppt ✓)
- Transportation in double bags under vacuum. Unpacking and tank loading from a glove box.
- **Batch production: 1 MS batch/day** (420 kg PPO + 504 g bis-MSB + 4 m^3 LAB)

Plant fully commissioned onsite!



Results:

- MS purification reduces ^{238}U/ ^{232}Th more than 1 order at ppq level
- In **LS:** **^{238}U<0,28 ppq; ^{232}Th<0,22 ppq** (preliminary results)


5000 m³ storage tank
Slope tunnel entrance
AFP plant
Mixing plant
Distillation plant

1,3 km SS pipe for LS
to the Underground LS Hall

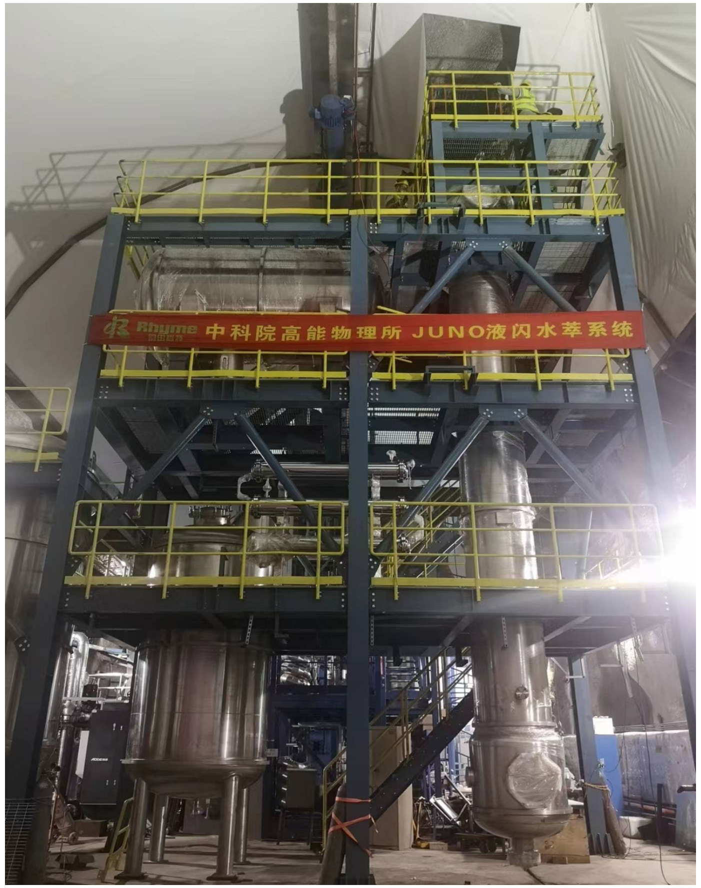
④ Water Extraction plant

Goal: removes polar contaminants and ions containing ^{238}U, ^{232}Th, ^{40}K

Underground LS hall

⇒ Radiochemical purification of LS

13m-high extraction tower with 30 turbine stages



Plant operation:

- LS and UPW stirred together; **LS** spread into **2-3 mm droplets**
- Counter-current flow: LS fed at the bottom (dispersed pahse), water at the top (continuous phase)
- **Custom shaft layout with 30 turbine stages**, separeted by perforated plates
- Double filtration (200 nm; 50 nm)

[Ye et al., NIM A, 10.1016/j.nima.2021.166251]

4 Water Extraction plant - results

Process parameters:

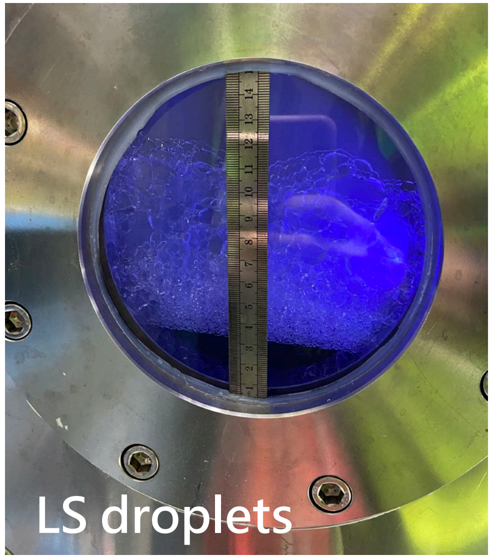
- LS:UPW ratio is 3:1 (7:2,3 m^3/h)
- T≤40°C preferred to avoid too much residual water left in LS
- N. theoretical stages: ≥5

Extraction tower parameters	
Water-LS ratio	1:3
Height	13 m
Diameter	1 m
N° turbine stages	30
Temperature	Up to 70°C
Rotation speed	30-60 r/min

Plant fully commissioned onsite!

Results:

- $^{238}U<0,28$ ppq; $^{232}Th<0,22$ ppq (preliminary results)
- Residual water content: 150-200 ppm (after filters)
- No worsening of absorption spectra



⑤ Gas stripping plant

Goal: - removes gaseous impurities ^{222}Rn, ^{85}Kr, ^{39}Ar, O_2
- removes **residual water** left in the LS after WE plant

Underground LS hall

⇒ Radiochemical purification of LS

9m-high stripping column, with unstructured packing

Column operation:

- Gaseous impurities dissolved in the LS transferred to the stripping gas by desorption mechanisms
- Counter-current flow: LS fed at the top by gravity, gas at the bottom
- Column filled with **unstructured packing (Pall rings)**, to increase the contact surface

Stripping column parameters	
Packing	Pall Rings, 13mm
Interface area	430 m^2/m^3
Height	9 m
Diameter	500 mm
Temperature	70°C
Pressure	250 mbar
N_2 flow rate	15 Nm^3/h

[Landini et al., NIM A, 10.1016/j.nima.2024.169887]

5 Gas stripping plant - results

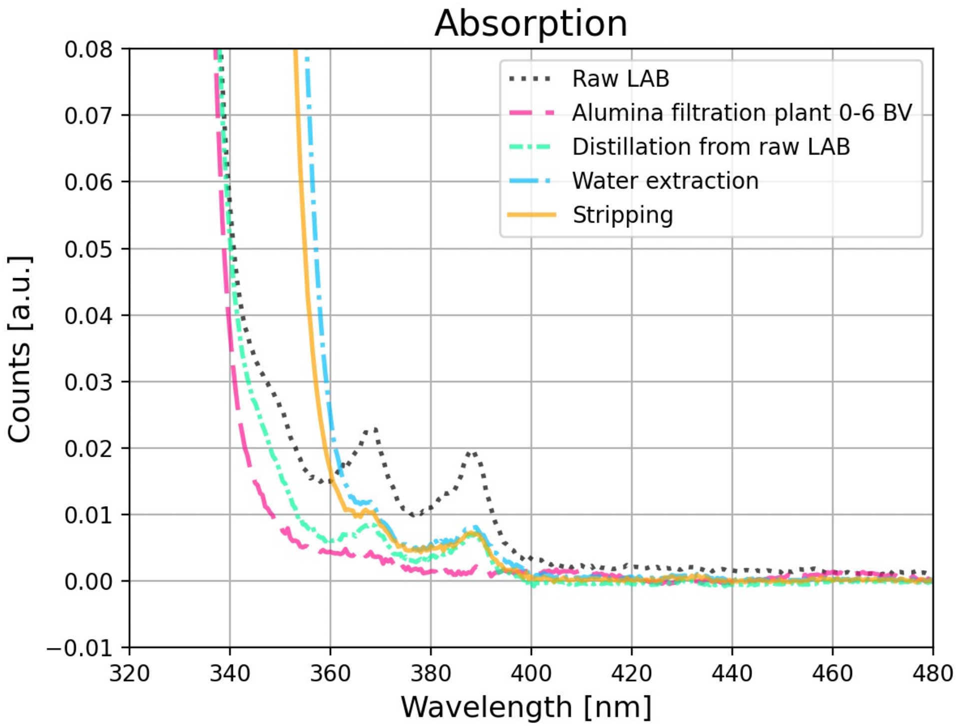
Process parameters:

- Stripping under vacuum: **250 mbar, 70°C**
- Stripping gas: **15 Nm^3/h** nitrogen (**HPN**)
- N. theoretical stages: 3-4
- Expected efficiency for Rn: ~95%

Plant fully commissioned onsite!

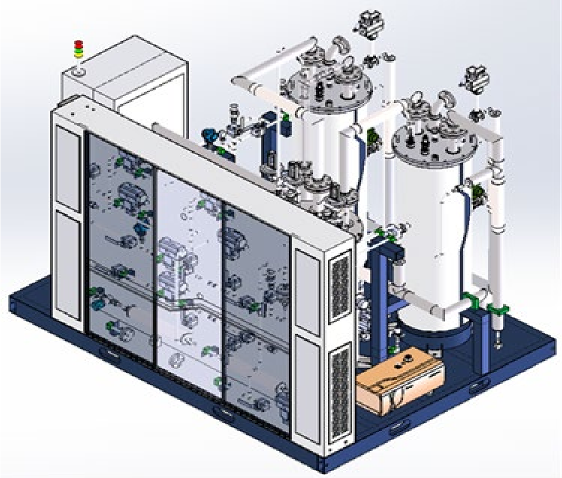
Results:

- $^{238}U<0{,}28$ ppq; $^{232}Th<0{,}22$ ppq; $^{40}K<0{,}7$ ppq (preliminary results)
- Water removal: from 150 ppm to **20 ppm**
- Excellent particle counting (much better than Class 50 MIL-STD-1246C)
- No worsening of absorption spectra



Auxiliary systems

High-purity nitrogen (HPN) system



- Low-temperature adsorption (LTA) technology
- Requirement: $^{222}Rn < 10\ \mu Bq/m^3$ - $^{85}Kr < 50\ \mu Bq/m^3$ - $^{39}Ar < 50\ \mu Bq/m^3$
- Flow rate: 100 Nm^3/h
- Usage: blanketing for plants, 5000m^3 tank and CD; stripping gas for stripping plant

Ultra-pure water (UPW) system

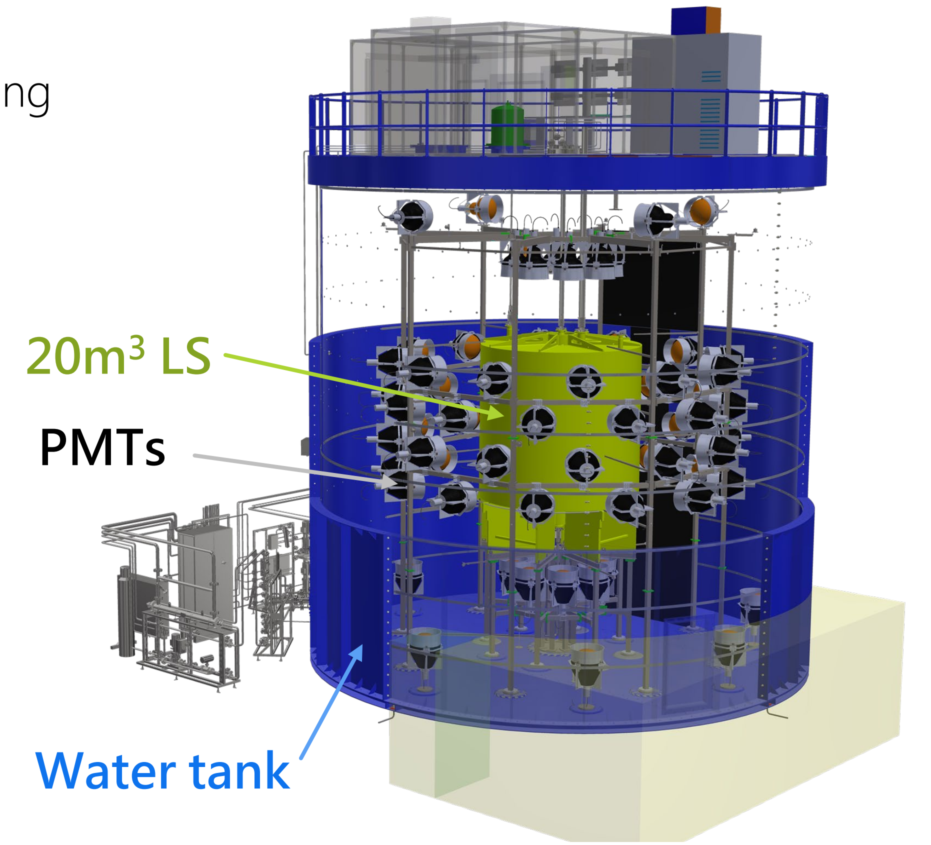
- Several devices (RO, degassing membranes, filters,…)
- Requirement: $Rn < 1\ mBq/m^3$ - $Ra < 3\ \mu Bq/m^3$ - $U/Th \leq 10^{-16}\ g/g$
- Flow rate: 2.5 m^3/h
- Usage: water extraction, OSIRIS

HPN and UPW are very important, since **they set the purification limit that can be reached in the purification processes** (WE, stripping)!

OSIRIS system

OSIRIS (Online Scintillator Internal Radioactivity Investigation System)

- Goal: **online measurements of the final LS radiopurity**, before filling into JUNO detector
  - Similar to CTF in Borexino
  - 1/7 (1 m^3) of the produced LS checked online during JUNO CD filling
- Design:
  - Cylindrical inner acrylic vessel (AV) with **20m^3 LS**
  - External **water tank** (550m^3, 3m water shielding against external background)
  - 64+12 20"-PMTs
  - Calibration (γ sources, laser)
- Sensitivity goal: $10^{-15}/10^{-16}$ g/g for U and Th



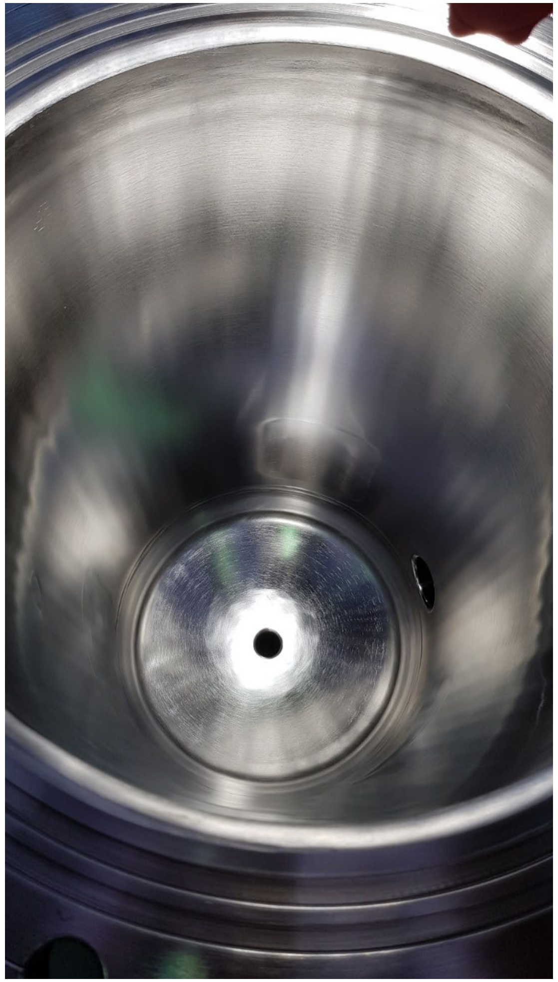
In case of unqualified LS samples, the LS can be sent back and re-purified by WE and Stripping plants.

[JUNO coll., EPJC, 10.1140/epjc/s10052-021-09544-4]

Common features

Cleaning procedure

- Material selection: SS 316L or 304L
- Special **cleaning procedure** for plant **internal surfaces**:
  1. Mechanical polishing (roughness≤0.4 um)
  2. Orbital welding and local pickling
  3. Degreasing
  4. Electro-polishing
  5. Mild Passivation, with 25% nitric acid + 75% demineralized water
  6. Water rinsing (2 cycles; until pH>5)
  7. Filtered Compressed Air Drying
  8. Quality check (particle counting, pH and conductivity tests, roughness measurement, endoscopic inspection)
  9. Final water rinsing after installation at JUNO site



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Plant leak tightness

It is important to prevent air and external contamination to enter the plants and come in contact with LAB/LS, to avoid pollution and oxidation.

- JUNO leak tightness requirements:
  - Single leak rate: <10^{-8} mbar L/s
  - Integral leak rate: <10^{-6} mbar L/s
  - **Plants sealed and leak-tested** through plant-wide campaigns

- **Flange design** with double oring protection and purging ports for HPN flushing

- Gas **blanketing** of all tanks, columns and equipment using **HPN**

QA/QC methods for LS

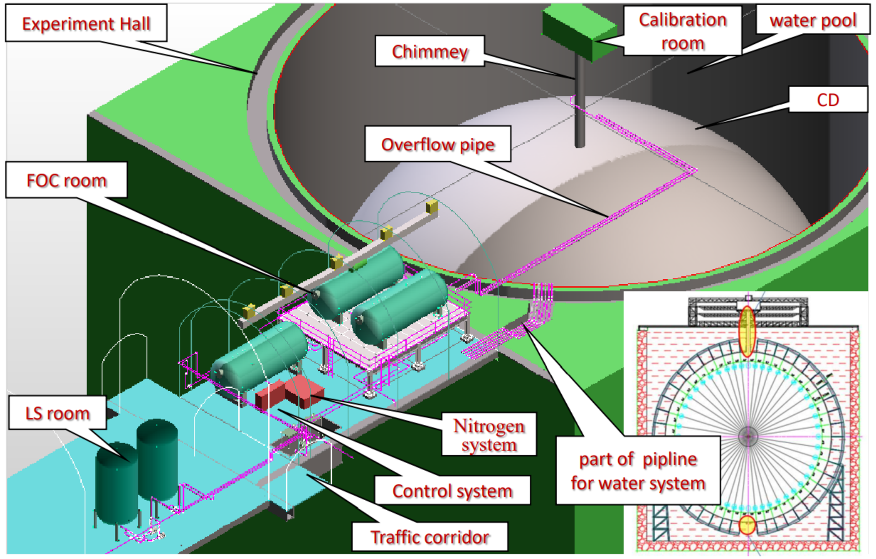
LAB/LS samples analysed through **several QA/QC methods**, selected and **optimized with dedicated procedures** to achieve better detection limits and sensitivity.

- For optical features:
  - **Absorption spectra** (spectrophotometer)
  - **Emission spectra** (spectrofluorometer)
  - **Attenuation length** (custom device: 3m tube with CCD camera)
  - Fluorescence time profile (custom setup with PMTs)
  - Refractive index (ellipsometer, refractometer)
  - Light yield (custom setup with PMTs)
  - Aging test
- For radiopurity:
  - **NAA** (Neutron Activation Analysis) (for U,Th,K) → see Andrea Barresi's talk
  - **ICP-MS** with pre-concentration (for U,Th,Pb)
  - Rn measurement system (in HPN and LS)
  - ^{14}C screening in LS → see Mingxia Sun's talk
- Others:
  - **Particle counting**
  - Device for water content in LS
  - Device for LAB/LS content in water

Detector filling

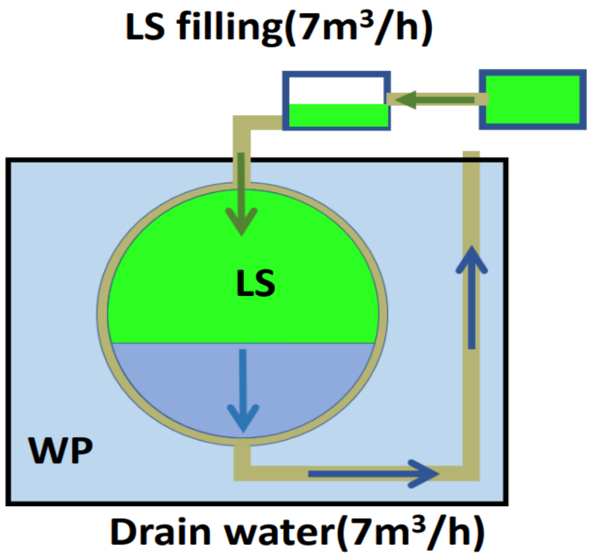
FOC system (Filling-Overflow-Circulation) is responsible for the filling operations of the central detector.

- Water filling of CD and water pool
- LS-water exchange scheme inside the CD



Schedule:

- Commissioning phase of the purification plants almost completed
- **LS filling** of the detector foreseen to start in **early 2025**
- **6-months** continuous filling (24/7) at $7m^3/h$



Summary

- Purification of the liquid scintillator is mandatory to meet the JUNO optical and radiopurity requirements.
- **5 purification plants for JUNO**: AFP, vacuum distillation, mixing plant, WE, gas stripping.
  Auxiliary systems suppling UPW and HPN.
  OSIRIS system for LS online monitoring.
- Material selection and screening, internal surface cleaning and leak tightness are crucial points.
- **Several quality control methods**, relying on state-of-the-art and/or custom devices.
- **Promising preliminary results** from the purification plants commissioning onsite.
- JUNO LS filling foreseen to start in early 2025.

Thank you for your attention.
Questions?