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“Investigations of the characteristics of installation for the measurement of low activities of 37Ar based on the detection of liquid argon scintillation"

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Argon-37 in subsoil air

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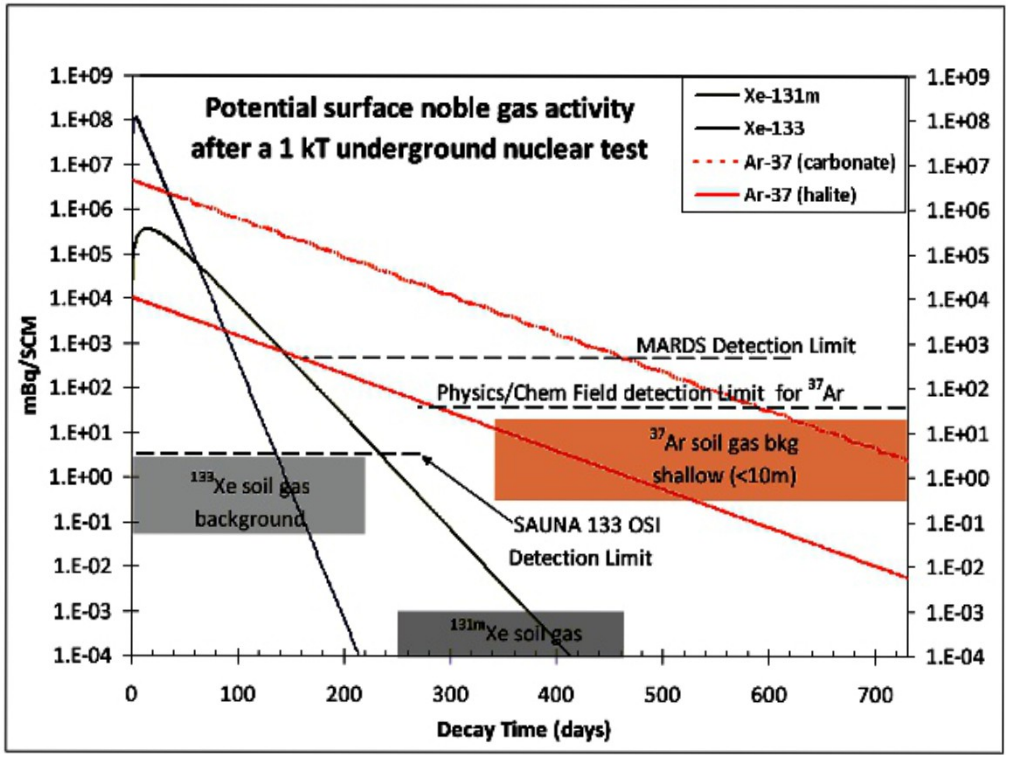


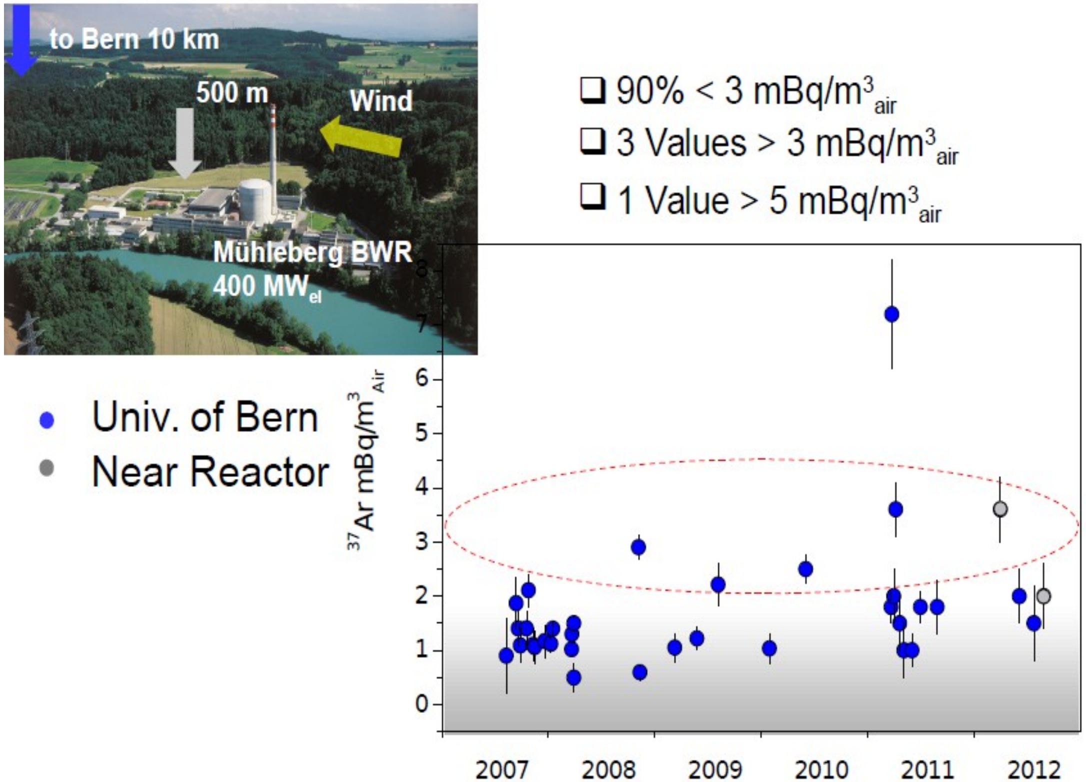
Pic. 1. Subsoil concentration of 37Ar, 131mXe and 133Xe after explosion of 1 kt. /1 /

One of the most significant evidence of a violation of the CTBT is the detection in the subsoil air elevated concentrations of the argon-37 which is formed in large quantities during the interaction of a nuclear explosion neutrons with calcium, which is a part of the rocks.

The concentration of argon-37 retains high values for much longer time than xenon radionuclides, so it is the "ideal witness" of a nuclear test /pic.1/.

/1/ D.A. Haas, H.S. Miley, J.L. Orrell, C.E. Aalseth, T.W. Bowyer, J.C. Hayes, J.I. McIntyre. The Science Case for 37Ar as a Monitor for Underground Nuclear Explosions. Report PNNL-19458, June 2010.

Argon-37 in atmospheric air



Pic. 2. Atmospheric argon-37 concentrations measured near Bern in 2007 – 2012 years /2/.

/2/ Roland Purtschert, Robin Riedmann, Lauren Raghoo. Ar-37 in atmospheric and sub-soil gases. 2014 Safeguards Symposium, Vienna.

The local background concentrations of 37Ar in the air depends on the location (proximity of nuclear facilities). The long-time measurements near Bern in 2007 – 2012 years show a variation from 0.4 to 3 mBq /m3.

Registration of a concentration of more than 5 mBq/m3 may indicate the fact of high neutron emission into the atmosphere.

Methods of argon-37 measurements

Traditionally, the measurements of ^{37}Ar are carried out in the underground laboratory of Bern (Fig. 3). Later, a low background counter was designed in PNNL, USA, (Fig. 4). Lately, a mobile system MARDS-II, intended for OSI, was also developed (Fig. 5).

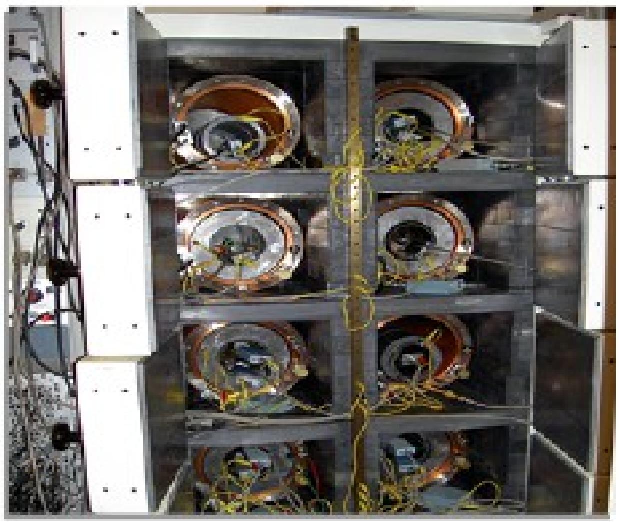


Fig.3. Proportional counter in active shield in Bern /2/.

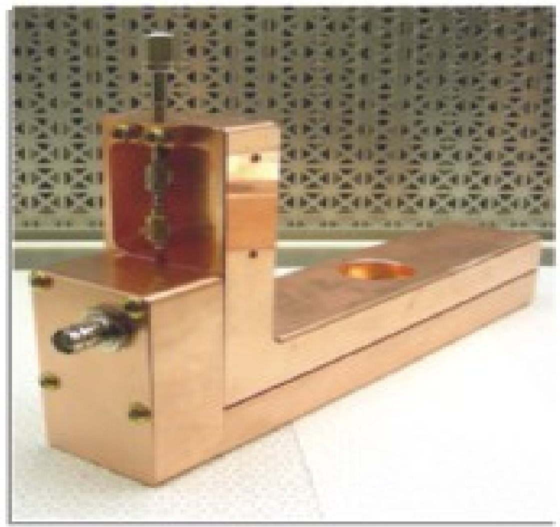


Fig.4. Proportional counter of 100 cc volume in PNNL /2/.

Fig.5. Equipment of mobile laboratory Mards-2. /3/.

/3/ LI Wei, YAN Zhaotong, XIANG Yongchun, WANG Hongxia, XIANG Qingpei, ZE Rende, LIU Qiang, GONG Jian. Latest Development on MARDS-2 an Argon-37 Detection System. Institute of Nuclear Physics and Chemistry, China Academy of Engineering Physics. SnT2015 Vienna.

Liquid scintillation method of argon-37 measurements

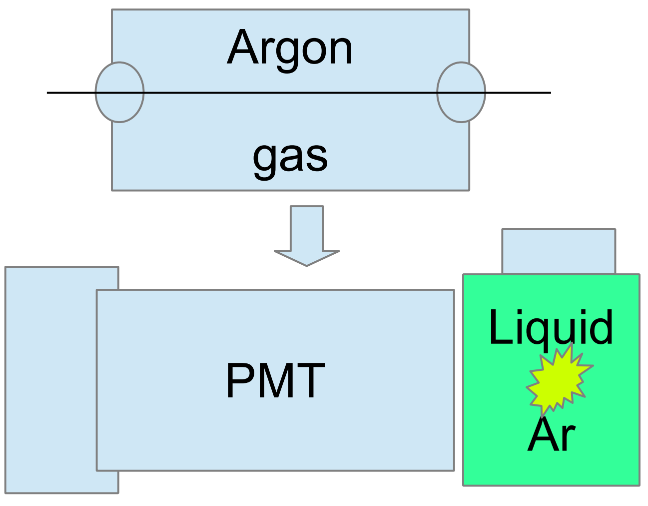


Fig.6. Instead of gaseous samples of argon to liquefied

The volume of the proportional counter cannot be significantly increased. Due to the high content of argon in the atmosphere, the volume of equivalent air sample also cannot be significantly increased, which imposes a limit on the sensitivity and accuracy of the method.

One of the possible solutions to the problem of increasing the sample volume is to convert argon to a liquid state. Due to the good scintillation properties of liquefied noble gases and, in particular, argon (Table 1), it is possible to measure the activity of argon-37 in the sample by registering directly the scintillation of the liquid argon itself /4, 5/

/4/ S.A. Pakhomov, Yu. V. Dubasov. Installation for measurement of low argon-37 activity (project). CTBTO International conference: Science and Technology, Hofburg Palace, Vienna, Austria, June 17-21, 2013.

/5/ S.A. Pakhomov. Prototype of laboratory system for detecting of argon-37 scintillations in liquefied samples of atmospheric argon. International Noble Gas Experiment Workshop 2017, 27 November – 1 December. National Physical Laboratory (NPL), Middlesex, UK

Argon-37 radioactivity

Table 1/ PRINCIPAL RADIATIONS PRODUCED IN THE DECAY OF ^{37}Ar

Decay Mode	Percent of All Decays	Energy of Auger Electrons (keV)	X-ray (keV)
K	81.5	2.823	0.0
L.............	8.9	0.270	0.0
K	2.7	0.202	2.621
K	5.5	0.201	2.622
M............	0.9	0.018	0.0
K	0.5	0.007	2.816

Ar-37 ($T_{1/2}$=35.04 d) decays by electron capture (EC) with emission of Aguar electrons with very small energy of 2.8 keV.

The wavelength of the light emitted in LAr (128 nm) is in the far vacuum ultraviolet (VUV) region and its direct detection requires special PMT with MgF2 windows or use of wavelength shifter - TPB – 1,4,4-tetraphenyl-1,3-butadiene.

The yield of scintillation light of liquid argon (40 phot/keV) somewhat exceeds even the output of NaI (38) and in 4 times exceeds the output of organic scintillators.

Auger electrons of argon-37 produce a high ionization density, which leads to a significant decrease in light output - up to 30 - 50% of the initial one due to ionization quenching.

Фазовые диаграммы аргона и азота

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One of the critical moments is the narrow temperature range for the existence of argon in the liquid phase at atmospheric pressure: from -185.89 to - 189.26 °C (Fig. 8). Given that the temperature of liquid nitrogen at atmospheric pressure is -195.80 °C (Fig. 7), which is 6.54 °C lower than the solidification temperature of argon.

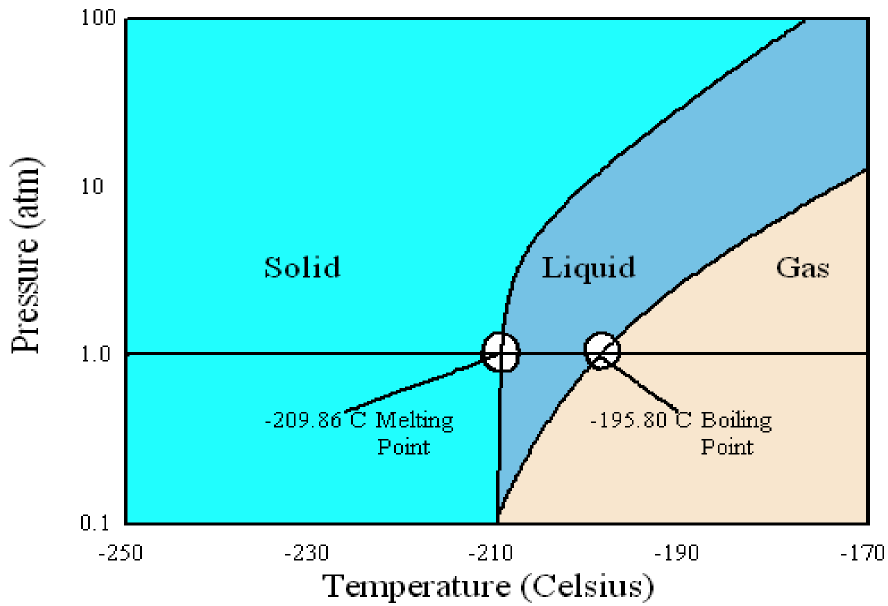

Рис. 7. Фазовая диаграммы азота

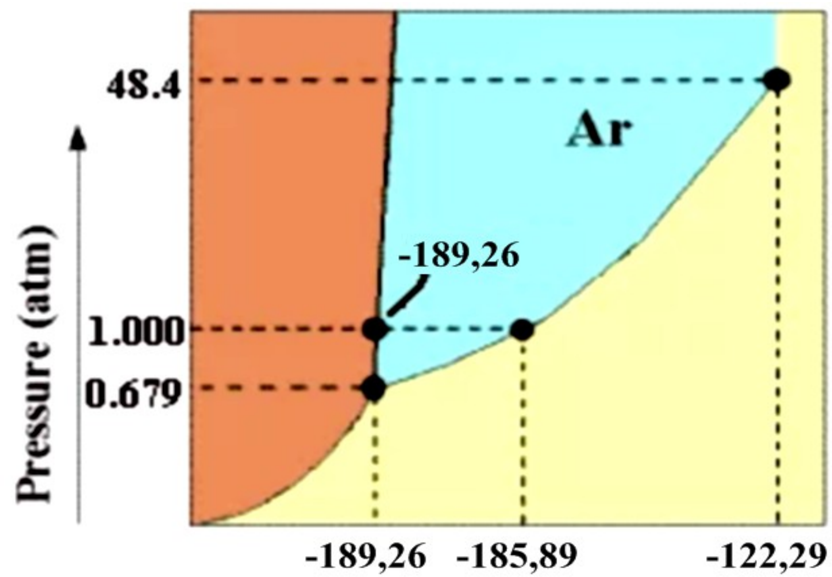

Рис. 8. Фазовая диаграммы аргона

Adjusting the temperature of a sample of liquid argon in a range of about three degrees wide is a difficult technical task. However, if the pressure in the measuring chamber is increased by 1.5 bar (bringing the absolute pressure to 2.5 bar), the control range can be extended to 10 degrees.

The installation for registration of liquid argon scintillations

The functional diagram of the installation for registration of liquid argon scintillations is shown in Fig. 9 installation for recording liquid argon

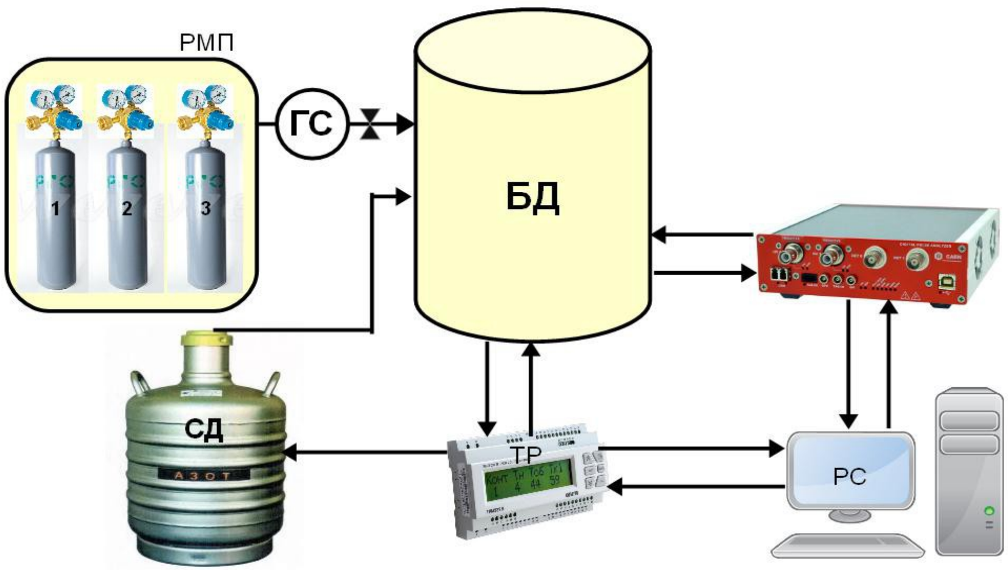

Fig.9. The functional diagram of the installation

Preprepared gases and gas mixtures are in cylinders 1-3 with a capacity of 10 liters under a pressure of 50 bar, mounted on the ramp. Cylinder 1 contains highly pure argon of brand 6.0 (background gas), cylinder 2 contains argon with the addition of 37Ar (with a volume activity of ~ 100 Bq per liter) and cylinder 3 contains the same gas, but with the addition of pure xenon of brand 5.0 to a volume content of 1%). Using a simple gas system, the gas from the cylinders under a small excess pressure of ~ 1.5 bar enters the detecting unit.

The detection unit

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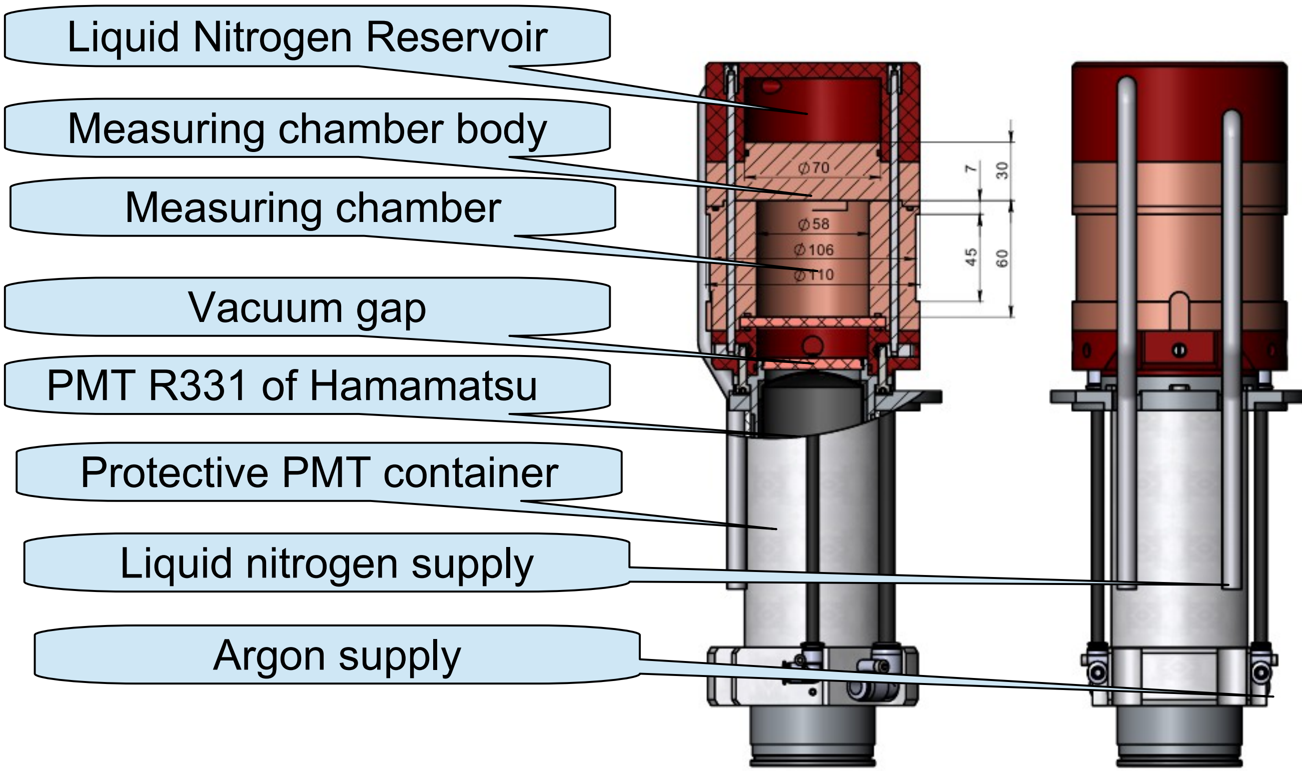

Fig. 10..The detection unit

The detection unit consists of a measuring chamber, the body of which is made of electrolytically purified copper and a Hamamatsu photo electron multiplier R331.

The PMT equipped with transparent windows, the volume between them is evacuated to protect the PMT from low temperatures.

The detecting unit is equipped with tubes for supplying liquid nitrogen and argon gas.

It is also equipped with temperature sensors and heaters to provide the temperature control.

The detection unit in shields

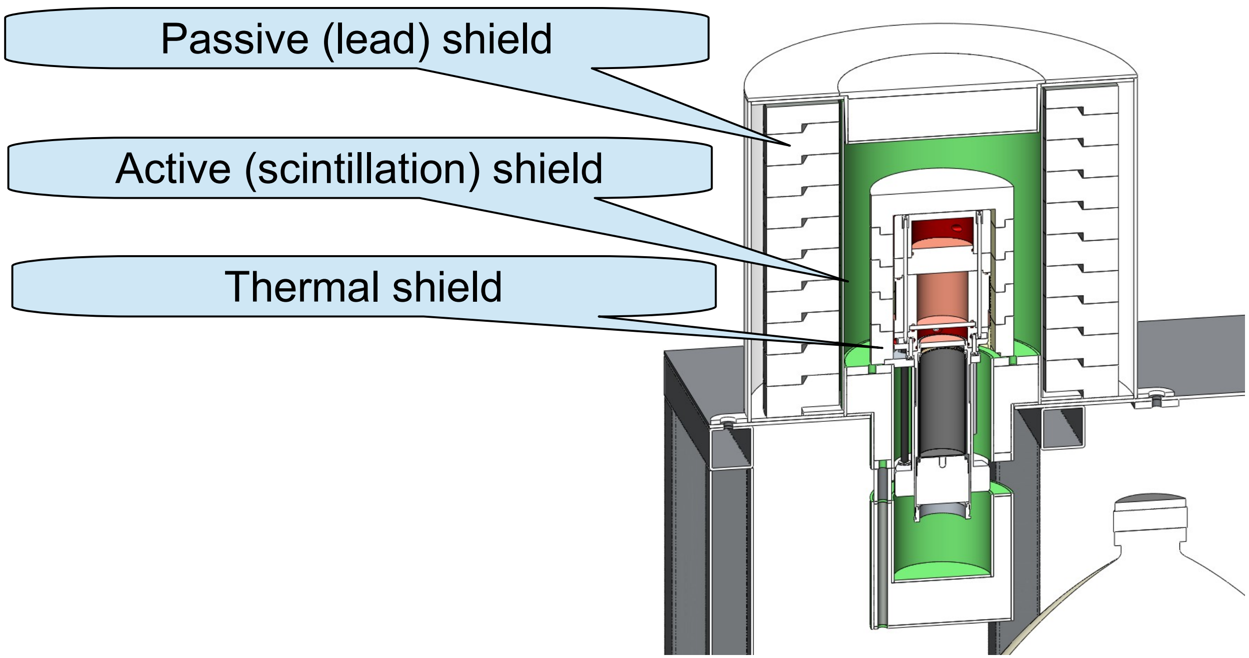

Fig. 11. The detection unit in shields.

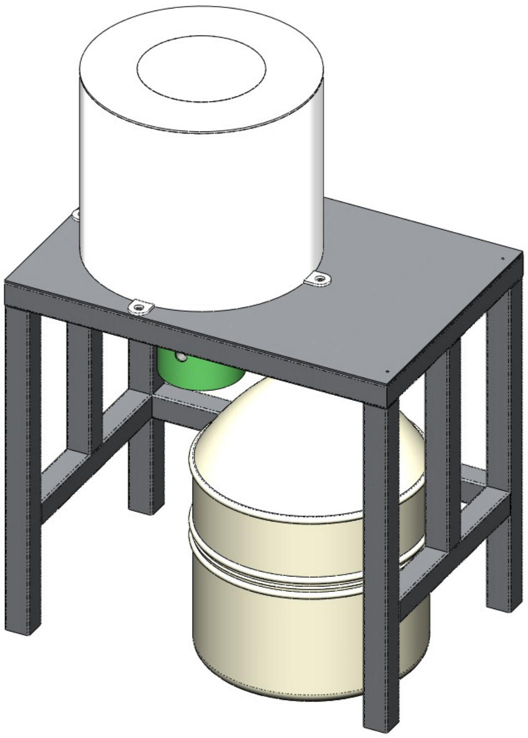

Fig. 12. The general view of detection unit

The technical requirements

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The technical specifications of the system must meet the requirements:

- The Minimum Detection Concentration (MDC) of at least 8 mBq/m3 air has to be achieved. The following criteria have to be met:
- The active volume of liquid argon shall be around 100 cm3
- Detection of at least 5 photo-electrons per decay of Ar-37
- Light collection efficiency of at least 70%
- Use of photomultiplier tubes with a quantum efficiency of at least 20%
- Investigation and possible use of wave-length shifters like xenon or tetraphenyl butadiene with nearly 100% shifting efficiency
- Oxygen and nitrogen contamination shall be in the order of tens of ppm at maximum
- The background count rate shall be 0.1 counts/second at maximum.
- Either active or passive shielding or a combination of both can be deployed
- Use of electrolytically refined copper and aluminum parts

PMT R331-05

The optimal choice of a photomultiplier for building argon-37 scintillations in liquid argon installations would be a low-temperature photomultiplier with a window that is transparent to ultraviolet radiation at a wavelength of 127 nm, which provides high photon detection. However, attempts to acquire such a photomultiplier were not successful, and we chose the Hamamatsu PMT R331-05 as an affordable alternative.

Tab.2. Characteristics of the R331-05

Parameter		R331	R331-05	Unit
Spectral Response		160 to 650	300 to 650	nm
Wavelength of Maximum Response		420		nm
Photo-cathode	Material	Bialkali		—
	Minimum Useful Area	46		mm dia.
Window Material	Material	Synthetic silica (Frosted)	Low K content borosilicate glass (Frosted)	—
	Shape	Concave-Convex		—
Dynode	Structure	Linear focused		—
	Number of Stages	12		—
Direct Interelectrode Capacitances	Anode to Last Dynode	2		pF
	Anode to All Other Electrodes	2.5		pF
Base		21-pin glass base		—
Weight		151		g
Suitable Socket		E678-21A (Supplied)		—

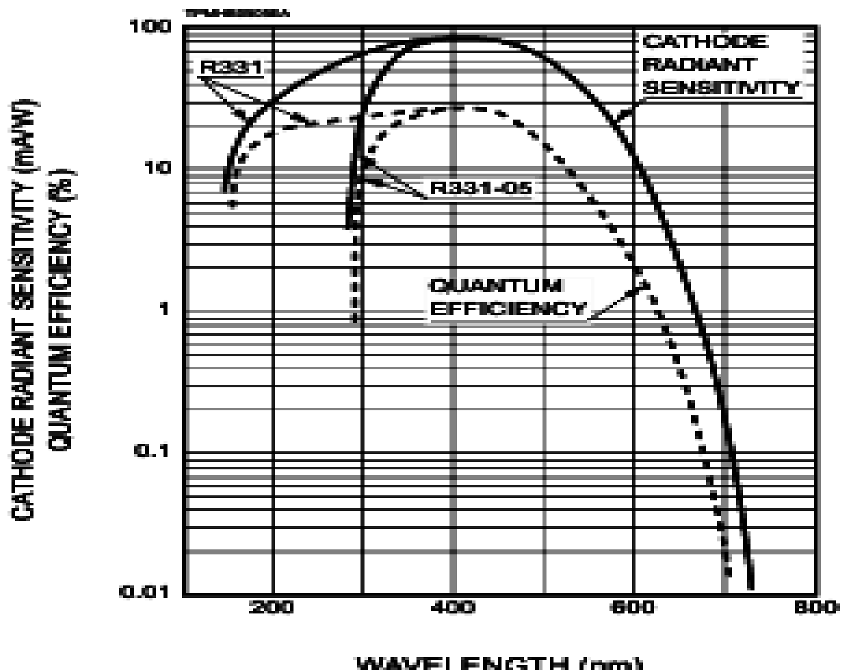

Fig. 12. Spectral characteristics of the R331-05

Expected installation counting characteristics

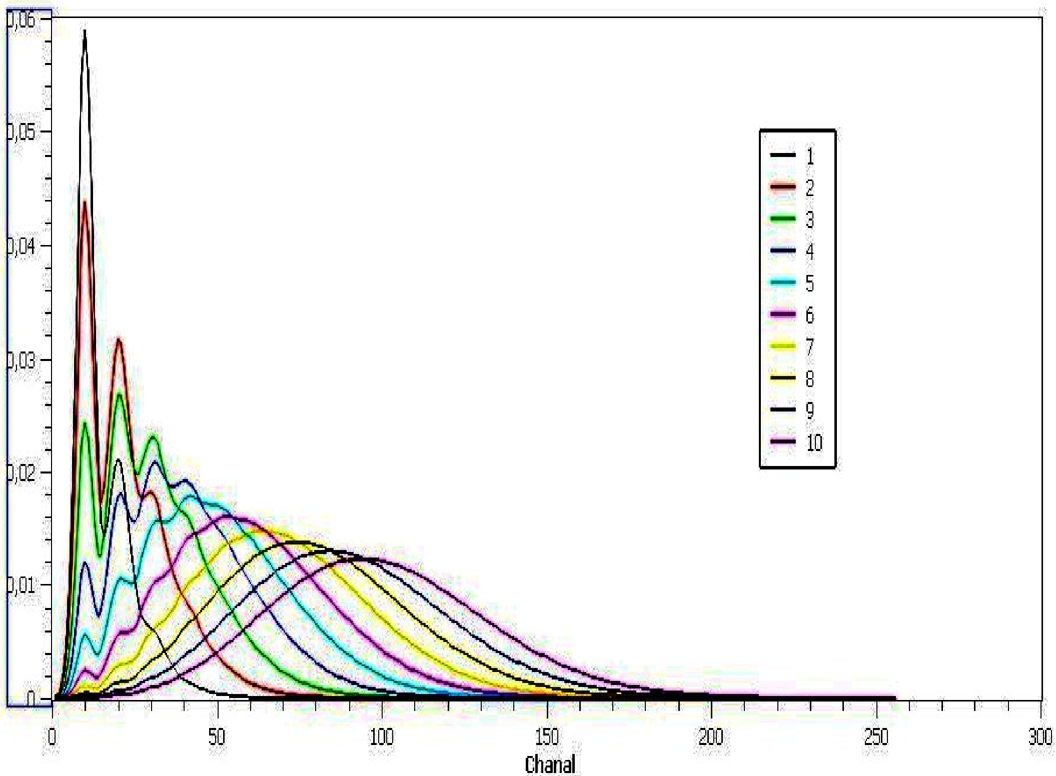

Fig. 13. Modeling of argon-37 decays spectrum depending on the number of photoelectrons.

The number of photoelectrons emitted by a photocathode of a photomultiplier when recording argon scintillations37 excited in liquid argon is: electron energy (2.8 keV) x light output (13.5 photons / keV) x light collection efficiency (75%) x photon wavelength conversion efficiency (100 %) x quantum efficiency of a photomultiplier (25%) = 7 photoelectrons per decay event.

The estimates show that the average number of photoelectrons> 5 corresponds to a registration efficiency of> 75%.

Conclusions

- At present, the work on the installation is not yet completed (in particular, the Caen analyzer has not yet been delivered).
- The main components of the detection unit, including a measuring chamber with a volume of 100 cm3, manufactured.
- According to the revised plan, the study of the system characteristics will be carried out in the autumn.
- In accordance with the updated estimations, the installation will satisfy the required characteristics.

Thank you