

Ultra high level liquid argon purification using electron emission from a tip array

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An array of 148 sharp tips was used to inject large quantities of free electrons in liquid argon and to purify it at a level < 0.09 ppb in few hours. We also evaluated the gettering capability of the stainless steel surface and measured the $I-I'$ characteristic of the array.

1. Introduction

The purification of liquid noble gases via the electron capture mechanism has been used by several groups, their main differences being the method of production of free electrons in the liquid. Irradiation with a strong γ -ray source [1], discharge of a filament in gaseous phase [2], and production of sparks [3] were successfully experimented. Here, an array of sharp points, negatively biased, generated a large electronic current in the liquid argon (LAr), allowing to reach a very high degree of purity in the liquid (oxygen equivalent concentration < 0.1 ppb) in a few hours. Sharp tips had been already exploited for LAr purification [4], but with a final impurity concentration bigger by a factor 30.

We performed a systematic study of the LAr purity as a function of the injected charge and a rough evaluation of the total number of removable electronegative impurities was done. A simple model based on the basic theory of the electron capture by electronegative impurities was developed, but the measured purification efficiency does not agree with this model, being higher by about a factor 20 than the expected one.

2. The chamber

A scheme of the experimental setup is shown in fig. 1. A 1.8 l stainless steel chamber was located in a bath of LAr. On the top flange there was a stainless steel pipe for pumping, filling and emptying procedure: a cryogenic all metal valve separated the chamber from

the pipes at room temperature to prevent the contamination of the pure LAr due to their large outgassing. The total chamber volume is subdivided into three sections:

1) *Drift region.* A gridded ionization chamber defined a drift region of 11.5 cm. A series of guard rings were placed, 3 mm thickness, separated by 7 mm, to ensure electric field uniformity. Each ring, starting from the cathode, was connected to the following one by a $18 \text{ M}\Omega \pm 5\%$ carbon resistor. The grid, at 5 mm from the anode, was connected to the ground by a 39

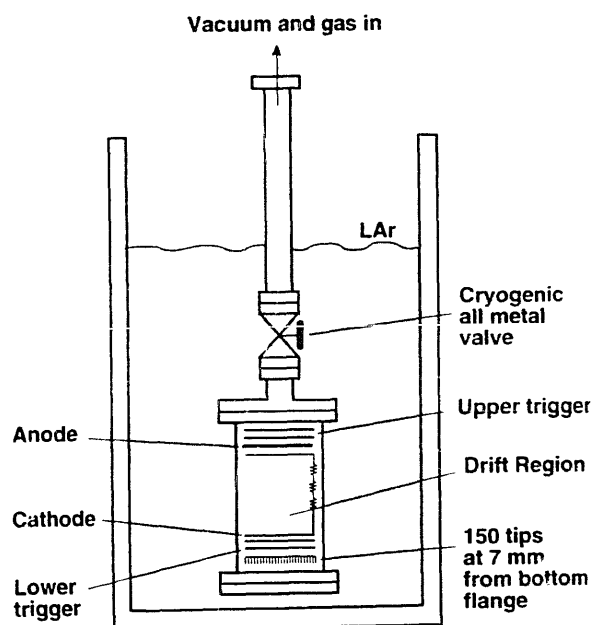


Fig. 1. Schematic view of the experimental setup.

$M\Omega \pm 5\%$ resistor. It was a copper electroformed mesh, 25 wires per inch, with geometrical transparency of about 98%. The field ratio $E_{\text{grid anode}}/E_{\text{cathode-grid}}$ was about 4, thus ensuring 100% transparency for the drifting electrons.

2) *Trigger system.* Two capacitors were placed above and below the drift region. Each of them consisted of two stainless steel disk, 5 cm diameter, 1 cm spacing. They acted as ionization chambers tagging cosmic rays which cross the drift region.

3) *Purification section.* An array of 148 sharp tips, made from a tungsten wire and mounted with regular spacing on a stainless steel disk, was placed under the lower trigger capacitor with the tips pointing to the CF100 bottom flange. The tips were obtained via electrochemical etching in a NaOH bath; their curvature radius was about $0.25 \mu\text{m}$ within 10%. All the tips, electrically connected together, had the same length and were at a distance of 7 mm from the bottom flange. The surface covered was about 12 cm^2 .

The stainless steel bottom flange was held at ground potential and acted as "gettering" anodic surface, whereas the tip array was biased at negative high voltage.

All the metallic electrodes were supported by Macor insulators. The procedure for cleaning the chamber components and the vessel itself was the same as described in ref. [5].

3. Electronic readout

The general layout of the electronic readout chain is shown in fig. 2.

A negative voltage of 2 kV was applied on the two inner plates of the trigger capacitors; the other plates

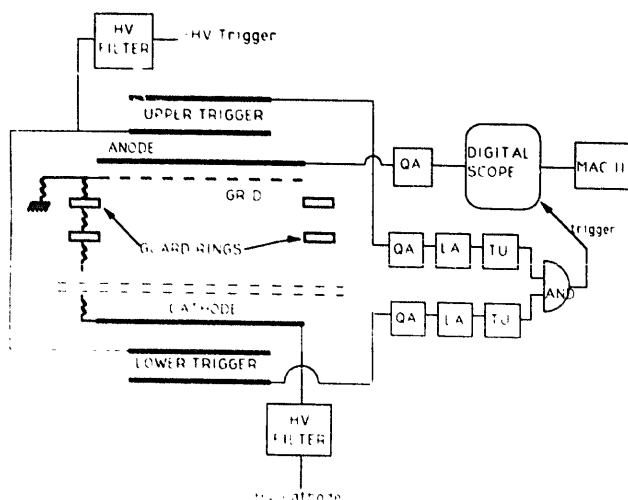


Fig. 2. Scheme of the electronics and read-out system. QA = charge preamplifier, LA = linear amplifier, TU = discriminator + timing unit.

were connected directly to the charge amplifier (QA). Each QA was followed by a linear amplifier (Laben 8340), a discriminator and a timing unit before entering in the coincidence unit. The AND output triggered directly the digital storage oscilloscope (HP 54502), read by a MacIntosh II computer via a GPIB interface.

The anode signal was sent to a charge preamplifier and read by the digital oscilloscope. The anode QA was chosen in order to have low noise and a high integration time τ_{ampl} to preserve the signals with long drift time. In our case we used two types of QAs: one with about $400 \mu\text{s}$ of integration time for low drift times and another with $\tau_{\text{ampl}} \approx 1.5 \text{ ms}$ for drift times of the order of ms.

The discrimination thresholds for the trigger signals were set at half of the amplitude expected for a minimum ionizing particle. The trigger rate was about 40 triggers/h. The spurious triggers were 2–3% and were discarded in the data scanning.

4. Data analysis

The electron lifetime τ was measured by fitting the ionization charge signal $Q(t)$ of the cosmic rays crossing the whole drift region. The fit procedure is well described in refs. [1,5], while the off-line analysis was the same as in ref. [1].

When the electron lifetime is less or equal to the preamplifier integration time τ_{ampl} , the fit is very sensitive. Fig. 3a shows the τ distribution for a run with $\langle \tau \rangle = 130 \mu\text{s}$ and $\tau_{\text{ampl}} = 400 \mu\text{s}$: the distribution is very sharp around its mean value.

For high values of τ the fit is less effective: with $\tau_{\text{ampl}} = 1.5 \text{ ms}$ we obtained distributions like in fig. 3b, where the long tail toward extremely high values was cut. In this case our measure gives only a lower limit for τ . On the other side, it was very difficult to use an amplifier with longer integration time because of the heavy low frequency background.

5. Experimental procedure

The vessel containing the chamber was baked under vacuum at 150°C for 24 h. A leak test was performed at room and LAr temperature with a sensitivity of 10^{-9} mbar l/s. The bottom flange, which acted as gettering surface, was activated under vacuum at 300°C for 24 h.

Once the chamber was cooled down at LAr temperature, it was filled with N60 gaseous Ar fluxed through an Oxisorb ^{#1} cartridge at a rate of 12 l/min. Not

^{#1} Messer Griesheim Industrial gas division, Dusseldorf, Germany.

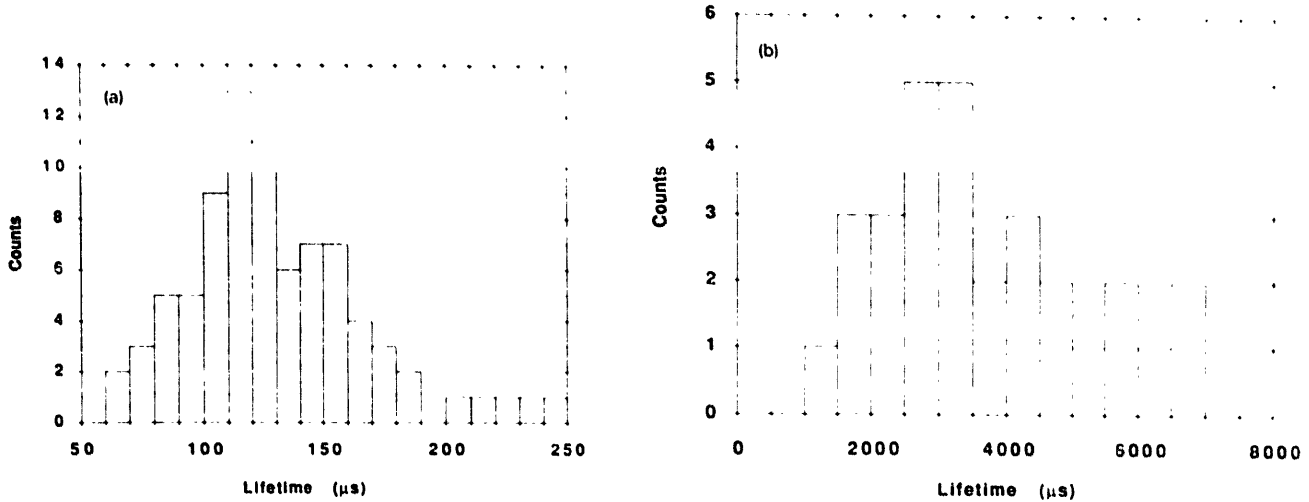


Fig. 3. Distribution of the electron lifetime τ for a run with $\langle\tau\rangle = (137 \pm 5) \mu\text{s}$ and $\tau_{\text{ampi}} = 400 \mu\text{s}$. (b) Distribution of the electron lifetime τ for a run with $\tau_{\text{ampi}} = 1.5 \text{ ms}$. About 30% of the entries have $\tau > 7 \text{ ms}$. With this cut, we obtain $\langle\tau\rangle = (3.4 \pm 0.6) \text{ ms}$.

much care was taken during this purification stage in order to obtain low purity level. The initial electron lifetime was measured; then we injected different amounts of electrons and after each injection τ was monitored. The injection current was typically hundreds μA and the injection time one or two hours. Once we reached a high purity level, we dirtied intentionally the LAR by opening the cryogenic valve. A second series of charge injection was performed to evaluate possible saturation effects of the gettering surface.

After each τ measurement, the I - V characteristic of the tip array was checked. The I - V behaviour was founded to be nearly parabolic, according to ref. [6], showing we were in the space-charge regime.

6. Results

The fig. 4 shows the electron lifetime τ vs the injected charge Q for the first series of data taking (immediately after the liquefaction and before opening the valve). The impressive result is that we need a charge of only 3 C (about 6 h with our HV supply) to reach $\tau \geq (3.4 \pm 0.6) \text{ ms}$ starting with $\tau = (57 \pm 1) \mu\text{s}$. τ is related to the impurity concentration n through

$$\tau = \frac{K(E)}{n},$$

where $K(E)$ is the attachment coefficient at the electric field E . With the value of K taken from ref. [7] we obtain

$$n_{\text{init}} = 5 \text{ ppb (ox. eq.)},$$

$$n_{\text{final}} \leq 0.09 \text{ ppb (ox. eq.)}.$$

The last charge injection did not seem to improve the electron lifetime. This is not due to a saturation effect of the gettering electrode, as we shall show later, but to the small sensitivity of the QA ($\tau > \tau_{\text{ampi}}$). Probably, the last lifetime value is in effect much higher.

The experiment τ vs Q values clearly exhibit an exponential behaviour, except for the last point for the reason previously given. It is possible to adduce a simple explanation. We assume that all the charged electronegative impurities drifting toward the gettering anode are definitively trapped there and the diffusion times are negligible with respect to the charge injection

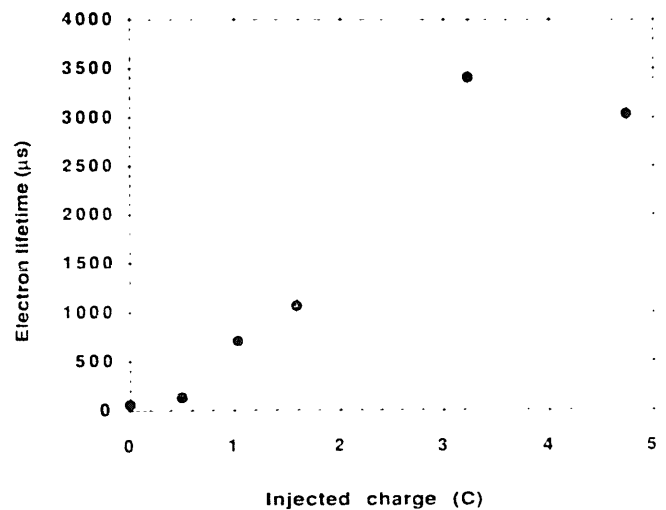


Fig. 4. Plot of the experimental τ vs Q values for the first acquisition series (after the liquefaction and before opening the valve).

times. Therefore the charge dQ required to remove a number dN of impurities is:

$$-dN = p \frac{dQ}{e},$$

where e is the electron charge, p the probability of attachment $= 1 - \exp[-T(E)/\tau]$, and $T(E)$ the tips to gettering anode drift time at the field E .

With a straightforward integration, we get

$$Q(\tau, \tau_0) = \frac{eN_0 K(E)}{T(E)} \ln \left(\frac{e^{T/\tau_0} - 1}{e^{T/\tau} - 1} \right), \quad (1)$$

where τ_0 and τ are the initial and final electron lifetimes and N_0 the total number of Ar atoms. For $T/\tau \ll 1$ as in our case, eq. (1) can be expressed as

$$\tau(Q, \tau_0) \cong \tau_0 \exp \left(\frac{T(E)}{eN_0 K(E)} Q \right).$$

Using the value of $K(E)$ from ref. [7] at the working electric field $E \cong 5$ kV/cm, we notice the expected curve is lower than the experimental one by a factor 20. We conclude that our simple model based on the electron capture mechanism is not adequate to explain the experimental behaviour of the mean electron lifetime versus the injected charge. Probably other phenomena contribute greatly to enhance the purification mechanism.

After opening the valve (elapsed time ≈ 30 min), the electron lifetime drops dramatically to (15.4 ± 0.3) μ s, corresponding to an impurity concentration $n = 20$ ppb (ox. eq.). The lifetime still grows exponentially with the injected charge but saturates at a value of about 400 μ s (see fig. 5). Our interpretation is that the gettering surface of the flange is completely saturated and cannot trap further impurities. We roughly estimate that the total number of impurities removed during the whole purification was 10^{18} molecules on a

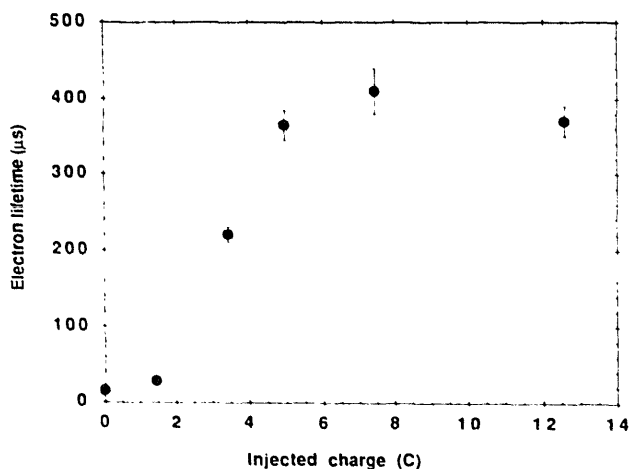


Fig. 5. Plot of the τ vs Q values after opening the cryogenic valve.

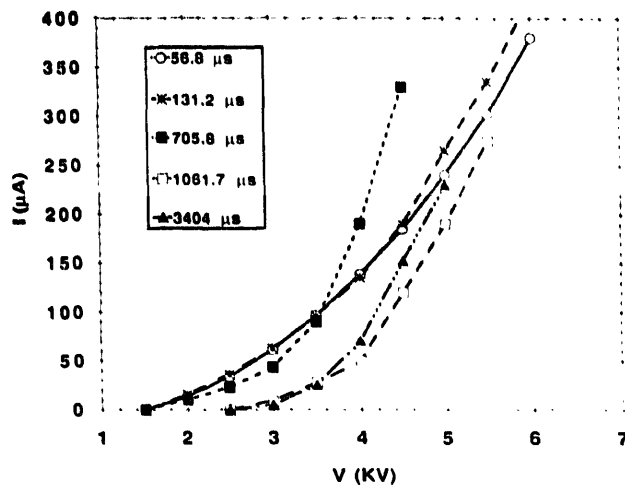


Fig. 6. Measured I - V characteristic of the tip array for different LAr impurity levels.

surface on 12 cm^2 . This value is in excellent agreement with the estimation of gettering capability of stainless steel (10^{17} molec/ cm^2) reported in ref. [1].

As mentioned above, the I - V characteristic of the tip array is nearly parabolic as shown in fig. 6:

$$I = \alpha (V - V_{\text{thr}})^2,$$

where α is a constant and V_{thr} is the threshold voltage. During the electron emission, both α and V_{thr} change with time because the tips become less sharp due to the bombarding action of the Ar positive ions. The smoothing of the tips, i.e. the increase of their curvature radius, shifts V_{thr} toward higher values and decreases α , as expected in the field emission. Hence, the increasing of α for curves with similar threshold may be interpreted as the dependence of the emission characteristic on the LAr purity. Moreover, with the same experimental setup and array but filling the chamber directly from the N60 argon bottle without the Oxisorb filter (impurity concentration of a few hundreds ppb), we noted that the emission current was lower of about two order of magnitude. This effect, if confirmed, could be a powerful method to monitor the liquid purity level.

7. Conclusions

The possibility to purify LAr in liquid phase at a level of several ms of electron lifetime in a very short time has been demonstrated. The only limitation of the method is the trapping capability of the gettering material surface. The purification system is very efficient starting from initial impurities of the order of tens of ppb. It is very useful as a second fine purification stage (for example, after the standard purification in gaseous phase) and to keep constant the purity level instead of a delicate recirculation device.

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