

Spark discharge method of liquid rare-gas purification

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The spark discharge method of liquid rare-gas purification is described. The method is sufficiently more simple than those widely used. Physical aspects of the method are discussed, and examples of its application are presented.

1. Introduction

One of the important requirements of liquid rare gases (LRG) to be used as a detector medium is a very high purity level. Because of high purity requirements every experimental facility in which detectors with LRG are used is supplied with a complex purification system.

Lately, a lots of work have been published about Ar purification and factors affecting the lifetime of electrons drifting over large distances in LAr [1–8].

It is important to notice that LAr is purified by more simple systems than LXe. It is well-known that there are difficulties in the purification of Xe when applying the same methods which successfully remove electro-negative impurities from LAr. Therefore the purification systems for LXe are even more complex.

As a rule, the purification system comprises an Oxisorb cartridge, a molecular sieve trap and a set of getters, each operating at its own high temperature.

The purity requirements restrict considerably the set of materials which can be used in the purification system and for chamber construction and demand thorough vacuum preparation [9].

The spark discharge method can essentially simplify the work with LRG. The method was worked out more than ten years ago and was primarily for the purification of LRG [10]. It was successfully used for purifying liquid Ar, Kr, Xe. The method allowed to get liquids with impurity concentrations less than 10^{-10} . And the limit was only determined by the sensitivity of the control method.

It is important to stress the fact that LXe was purified easily by this method since xenon purification is usually a rather difficult task.

By now the method has been used in several laboratories in the USSR for purifying in particular great volumes (hundreds of liters) of liquid [11]. Later the method was adapted for purifying rare gases in gas

state [12]. Now it is successfully used for purifying gas mixtures like Xe–H₂ [13].

The method is sufficiently more simple than widely used ones and to our opinion its possibilities are underestimated by some scientists and firms abroad.

2. Requirements for purification systems

The requirements for ionization detectors are well-known. For the centimeter detectors it is desirable to get an electron mean free path of about 10 cm and subsequently a lifetime of 50 μ s and impurity concentration of 0.5 ppb oxygen equivalent.

For scintillation detectors the purity requirements are less known. Here we can offer new results which brighten up the question.

The correlation of integrated ionization and scintillation pulses in a LXe chamber, during purification of the liquid with high initial impurity concentrations is shown in fig. 1. It can be seen that the scintillation pulse height stops to rise when the ionization pulse height equals only 1/10 of its maximum value.

In the chamber the relation between ionization pulse height A_i and electron lifetime, τ , is expressed by the equation:

$$A_i = A_0 \frac{\tau}{T} \left(1 - \exp\left(-\frac{\tau}{T}\right) \right),$$

where T is the electron drift time.

Under the condition $\tau/T \ll 1$ one can get $A_0/A_i \approx \tau/T$. So $\tau \approx 0.55 \mu$ s, $c \approx 45$ ppb.

The ratio of these two values (≈ 0.5 ppb for the ionization detectors and ≈ 50 ppb for scintillation ones) seems to be quite reasonable. Indeed the electron drift time through 1 cm is about 5 μ s, while the lifetime of excited states emitting the scintillation light is about 20 ns.

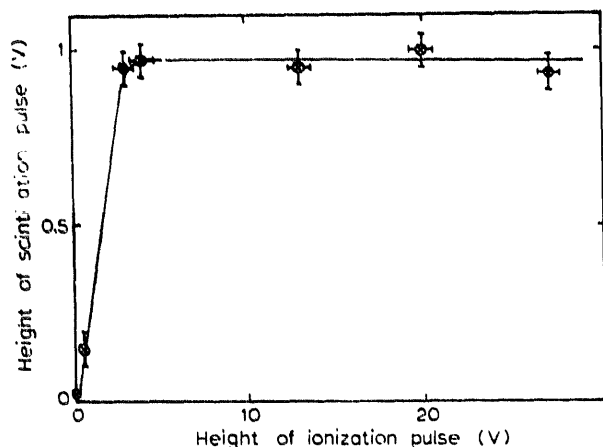


Fig. 1. Correlation between ionization and scintillation pulses in a LXe chamber during spark discharge purification which took place in the same chamber.

3. Physical aspects of the method

The principle of the method is as follows. Metal electrodes of special form are placed in the liquid and an electrical spark discharge is produced between them, in some way. As a rule the form of a discharge gap is a blade-plane. The following processes are responsible for purifying.

3.1. Disintegration of electrode material

It is well known that the disintegration of electrode material is widely used in the electro-erosion technology. Two main questions then arise, with competitive requirements. Productivity, which demands the largest energy in every pulse and quality of forming surface, which in the case of purifying means micro granularity, which demands small energy in every pulse. Some kind of electro-erosion technology allow to disperse metal with the velocity up to $15 \text{ cm}^3/\text{min}$. To get a good purification effect it is necessary to disperse metal dust grains not more than $1 \mu\text{m}$ each.

It is experimentally shown that the spark discharge effectively disperses the metal of the electrodes in a dense gas (with pressure of the tens of atmospheres). So the method can be used for purification in the gas state, in the gas storage reservoir particularly.

3.2. Adsorption of impurity molecules by metal dust on the chamber surfaces and "burying down" of adsorbed molecules by new portion of dust

These processes lie on the working principle of electrical discharge vacuum pumps.

The purification process can be described by the system of equations:

$$\begin{aligned} dN_x/dt &= A - kN_x N_m, \\ dN_m/dt &= -kN_x N_m, \end{aligned}$$

with initial conditions: $t = 0$, $N_x = 0$, $N_m = N_0$. Here: N_x , N_m are the numbers of active centers of adsorption and impurity molecules respectively, A is the velocity of creation of active centers, K the coefficient showing the probability of adsorption.

The solution of these equations in general gives a rather complex function. But at further stages of purification, after t_0 , when $N_x \gg N_m$, N_x can be considered as a constant and so we get a simpler expression:

$$N_m = N_m(t_0) \exp[-kN_x(t_0)(t - t_0)].$$

This expression agrees very well with experimental results which are shown in section 5.

3.3. Disintegration of complex molecules in discharge

3.4. Attachment of electrons to electronegative molecules and drift of negative ions

3.5. Effective liquid motion mixing up a liquid

It is quite important that spark breakdown leads to bubble formation and to intensive hydrodynamical motion. In non-optimal constructions, electrohydraulic blow can destroy insulators or even the walls of the chamber.

On the other hand, in purifying, liquid motion plays a positive role, mixing up a liquid effectively. To our mind, the spark gap in the corner of a big chamber with a long period of work (calorimeter, TPC etc.) helps purifying the liquid continuously without evaporating in the case of emergency.

4. Experimental technique

4.1. The preparation of the system

Of course it is better to prepare the system in the traditional way, but it might be difficult for some reason or another. Then the effectiveness of the spark discharge method allows to use a quite simple procedure, which takes no more than the short time to pump down to 10^{-2} Torr, and without baking. Thus the purity which is needed for an effective operation of a gridded chamber will be also achieved, in shorter.

For preliminary purification of gas we used the method of crystallization. Xenon was frozen in storage vessel up to 77 K (liquid N_2 temperature), then we pump out the impurity gas and after that we melt the xenon and repeat this procedure once again. The results can be seen in fig. 2.

4.2. Constructions of the purification chambers

Several variants of construction of spark gaps and many variants of purification chambers were studied

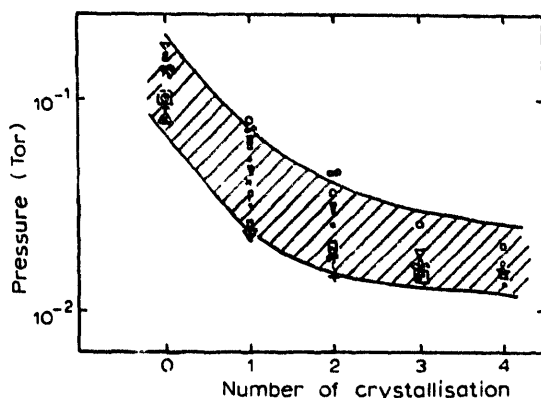


Fig. 2. The dependence of residual pressure above solid Xe at liquid N_2 temperature, on the number of crystallization. Various runs are shown.

for many years by using the spark discharge method of purification. As a rule, electrodes were placed into the same chamber of scintillation or ionization.

An example of ionization chamber with purification electrodes is shown in fig. 3. The control of purity by the analysis of form of current pulses, induced by short X-ray pulses was provided in the same chamber.

In scintillation chambers, spark electrodes were placed behind some shield and thus the chamber window wasn't dusted.

In our recent study two chambers connected by a tube, one for scintillation and one for purification, were put into one cryostat, which could rotate around a horizontal axis, and liquid Xe could pour over from one chamber into the other [14].

5. Results

The dependence of electron life time in LXe on the purification time is shown in fig. 4. It can be seen that the dependence is an exponential one. The velocity of purifying depends on the average current, linearly. The

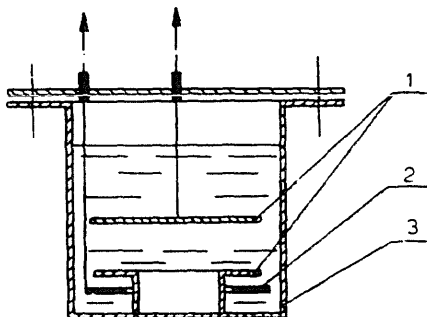


Fig. 3. Example of the ionization chamber with purification electrodes. (1) chamber electrodes, (2) disc blade, (3) wall of the chamber.

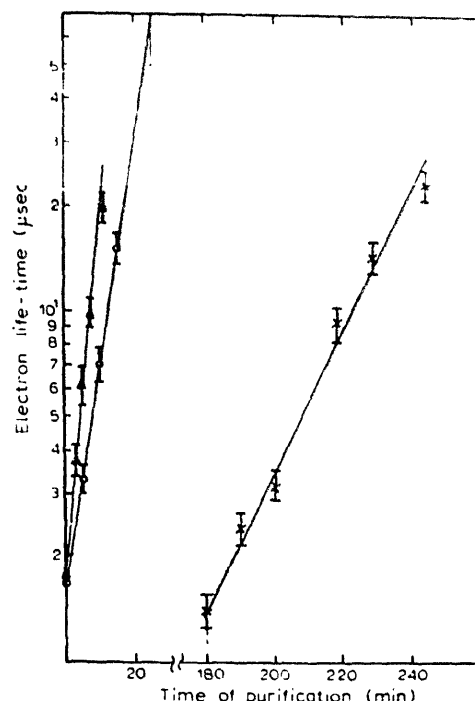


Fig. 4. Changing of free electron life-time during purification. (1, 2) gas after several crystallizations (see text), average current of spark breakdowns: (1) $I = 100 \mu A$; (2) $I = 60 \mu A$; (3) gas without preliminary crystallization, $I = 60 \mu A$.

velocity of purifying depends much on the initial impurity concentration.

The analysis of these dependencies shows that to remove one impurity molecule, about 100 atoms of Ti are needed, and the effectiveness of dispersing the Ti under the conditions of this experiment, is about $100 \mu g/C$. So the whole effectiveness of purification can be estimated as $10^{-16} C$ for impurity molecule. We used electrodes from various materials: stainless steel, aluminum, copper and titanium and did not find any difference in the velocity of purification. But in the case of titanium electrodes the layer of dispersed titanium had a large getter capacity and absorbed impurity molecules for hours after discharges stop.

It is useful to point out that in every purification chamber the sound of spark discharge can be heard and in the chamber with glass walls the color of spark discharge can be seen. During the purification, both sound and color are changed and the experienced investigator can determine the purity visually or by ear. The result will not be a pure scientific one but everybody can be taught to determine it this way, which is practically very useful.

It is also useful to point out that during this purification a breakdown voltage is being changed, namely it decreases. This is another indication for the experimenter.

6. Conclusion

The experience of application of the spark discharge method for purification of LRG, especially Xe, shows that it practically has no restrictions. To purify a certain volume of LXe a certain amount of Ti, will be used spending for it a certain amount of Coulombs. The duration of the purification procedure is determined by the average current which one can provide. To increase the current, one can increase the frequency of spark breakdowns or/and the number of independent gaps. For estimation one can use the following quantities: not more than 100 atoms of Ti for one impurity molecule. One spark gap ($C = 100$ pF, $U = 5$ kV) allows to disperse ≈ 100 μ g for Coulomb.

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