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Section A

A historical view on the R&D for liquid rare gas detectors

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A historical view on the R&D for liquid rare gas detectors starting from 1948 up to now is presented with some comments on their applications to future experiments.

1. Introduction

Investigation of liquid rare gas detectors was started in 1948, when the development of crystal detectors was facing a great difficulty, called the "polarization effect". Almost half a century has passed since then, during which much experience on liquid rare gas detectors has been accumulated. The present status of developing these detectors is in the stage where their applications to physics experiments in the field of elementary particle physics or space physics are being made. In this paper, the R&D for liquid rare-gas detectors made so far are reviewed, and comments based on the accumulated experiences are given for future development.

2. Dawning of liquid rare gas detectors

In the latter half of the 1940s, the development of the "crystal counter" was facing a technical difficulty, which was the so called "polarized effect". In 1948, by pointing out the absence of the "polarization effect" in rare gas liquids, Huchinson [1] and Davidson and Larsh [2] succeeded in observing conductivity pulses in liquid argon. In 1953, Marshall [3] constructed a liquid Ar gridded ionization chamber to measure high energy β-particles. This was the first application of a liquid rare gas detector to a physics experiment.

Following Marshall's experiment, Williams [4] and Sawan [5] carried out experiments to measure the electron drift velocity, ionization yield and electron–ion recombination in liquid Ar using the pulse technique. However, over the period from 1954 to 1967 very few researches were reported on the development of liquid detectors.

3. Research on fundamental properties of liquid rare gases as detection media

In 1968, Alvarez [6] proposed the use of rare gas liquids as a detection medium to realize position sensitive detectors with precise spatial resolution, and pointed out that liquid Xe would be the most promissing detection medium. He also pointed out a possibility of using liquid Ar as a detection medium in the calorimeters to be used in elementary particle physics experiment. Being based upon this proposal, the LBL group directed by Derenzo concentrated their effort on the R&D of the realization of a position sensitive detector using liquid Xe. They found that the electron multiplication in liquid Xe is the most stable one among the rare gas liquids [7], and obtained a good energy resolution of 48 keV FWHM for 279 keV γ-rays from a ^{203}Hg source [8]. The maximum gain of electron multiplication which they reported reaches ~ 200 times [8,9]. To realize a liquid Xe position sensitive detector, the LBL group constructed a liquid Xe multi-wire chamber [10] and found that when the wire spacing was reduced to less than 1 mm, the electron multiplication did not occur due to the reduction of the electric field strength near the wire. After this experience, they constructed another liquid Xe multi-wire chamber with a wire spacing of 2.8 mm as a medical imaging device [11]. They also constructed a one-dimensional microstrip liquid Ar chamber whose position resolution was reported to be better than 20 μm (rms) [12]. Since 1975, the LBL group has changed the direction of their R&D from the Xe detectors to the medical detector devices.

A Grenoble group [13] also found electron multiplication in liquid Xe, but slightly later than the LBL group. A Sacley group [14] constructed a liquid Xe gridded ionization chamber with a drift distance of 5

cm, which was the widest one ever made, and measured the energy spectra of γ-rays, yet the energy resolution obtained was unsatisfactory, being inferior to that of a NaI(Tl) counter [14]. However, I should emphasize the important work made by the Sacley group using the same chamber which is the discovery of "proportional scintillation" in liquid Xe [15], although they did not report the quantitative properties on proportional scintillation in liquid Xe.

In the end of the 1960s, a Waseda–Rikkyo–Riken collaboration group started the research on fundamental properties of liquid Ar and Xe as detector media, aiming at the application of the detectors based on these liquids to physics experiments [16]. The collaboration group initiated its work by measuring the *W*-values in liquid Ar and Xe by using the pulse technique, and obtained reliable data for both liquids purified by a Ba–Ti getter [17,18]. Using these *W*-values, the group estimated the ultimate energy resolution in liquid Ar and Xe ionization chambers, and the estimated values for both liquids were near that of Ge detectors rather than that of NaI(Tl) counters [19]. This had some impact on detector physicists, and interested them to participate in the R&D of the liquid Xe chamber. However, these estimated ideal energy resolutions have not been experimentally achieved yet [20]. The present status for understanding the mechanism that prevents us from achieving the ideal resolutions will be described later.

The group also investigated the mixing effects of Xe, N_2, and organic molecules into liquid Ar in terms of the ionization yield [21,22] and of the drift velocity of electrons [22]. They have also investigated the "proportional scintillation" as well as the electron multiplication process in liquid Xe [23,24], and have shown that the analysis method used for the proportional scintillation in the gas phase can be succesfully applied to that in the liquid phase as well [24].

According to the experience the group has obtained so far, the photon multiplication (i.e. "proportional scintillation") in liquid Xe is much more stable and technically less difficult to handle compared to the electron multiplication process. When the chamber is operated in the "proportional scintillation" region, we can simultaneously use charge signals as energy signals as well as the proportional scintillation signals, since the charge signal should be free from any deterioration of energy resolution in this region. From these points of view, it is my belief that the proportional scintillation technique will be useful for developing devices which can determine the incident position of ionizing radiation precisely.

The problem of obtaining the "ultimate energy resolution" in liquid Ar and Xe ionization chamber, which I mentioned before, still remains an unsettled issue. I would like to describe briefly the present status of the problem. The best value of the energy resolution for 1 MeV electrons experimentally obtained so far is 26 keV (FWHM) for the liquid Ar chamber [25] and 41 keV (FWHM) for the liquid Xe chamber [26]. These values are 7–22 times larger than the ideal values. To understand such descrepancies, Thomas [27] at CALTEC proposed a "δ-rays recombination model" which consists of the following assumption and fact:

i) A large fraction of ion–electron pairs recombine in soft δ-ray tracks or near the end of δ-rays due to the high ion–electron density.

ii) The number of such soft δ-rays is subjected to a statistical fluctuation.

The overall effect of i) and ii) results in a large fluctuation of the charge collected on the anode. In rare gases, the recombination occurs between an electron and a dimer ion and one recombination produces one UV photon. If the above model is correct, the doping of organic molecules, which can be ionized by recombination photons with a high quantum efficiency in liquid Ar or Xe, should significantly improve the energy resolution. In 1987, attending the workshop on liquid rare gas detectors held at UCLA, I proposed to verify experimentally whether the energy resolution in liquid Ar or Xe doped with photo-sensitive molecules is improved as predicted. Recently the Waseda group [26] has measured the energy resolutions for γ-rays and conversion electrons from a ^{207}Bi source using liquid Xe doped with TEA that has the highest photoionization quantum efficiency (~ 0.8 [28]) among various combinations of organic dopants and liquid Ar or Xe. The result clearly shows that the energy resolution is remarkably improved, especially at low electric fields. However, the best energy resolution obtained in this mixture is still far from that of the ideal case. Such an experimental result indicates that the Thomas model may be insufficient to explain why the experimental achievable energy resolution is always worse than the theoretically predicted value. Shibamura [29] has made the full comparison between the Thomas model and the experimental result.

4. Appearance of the liquid Ar sampling calorimeter

In 1974, three prototype chambers for the liquid Ar sampling calorimeter were constructed by Knies and Neuffer [30], Engler et al. [31] and Willis and Radeka [32] independently and were successfully operated. These prototypes were of the multi-paraliel-plates type and some parts of the plates were used as convertors. The calorimeter, having a simple structure, provided a simple calibration procedure and good time stability. Because of these advantages, this detector technique has been continuously employed for 4π calorimeters in many collider experiments. This is the first example of

successful application of rare gas liquid detectors to real physics experiments. I will skip the details of the development of liquid Ar sampling calorimeters, since the main subject of this paper is to review the R&D of liquid rare gas detectors. Instead, I would like to describe new ideas appearing in this field since 1987, and would like to discuss the possible application of these new ideas to physics experiments in the following section.

5. Proposals of new devices and their applications

Since 1977, many new devices based upon rare gas liquids have been proposed in the field of elemental particles or astrophysics experiment. These devices could be divided into following four categories:

1) Liquid Ar or Xe time projection chamber (TPC).
2) Homogeneous liquid rare gas calorimeter.
3) Scintillating liquid rare gas detector.
4) Photoionization detector.

5.1. Liquid Ar or Xe TPC

In 1977, Rubbia [33] at CERN proposed to construct a large scale liquid argon TPC for rare event experiments. At almost the same time, Chen and Lathrop [34] at UC Irvine also presented the idea of liquid Ar TPC. These goals were to detect solar or cosmic neutrinos or proton decay using liquid Ar TPC.

In realizing a large scale liquid Ar TPC, extremely high purity liquid Ar, in which the attenuation length of drifting electrons is expected to be longer than 1 m, is needed. In 1985 Aprile, Giboni and Rubbia [35] succeeded in obtaining such long attenuation lengths of electrons in liquid Ar purified by simple purification system. Having obtained this encouraging result, Rubbia proposed the construction of a 6 kton liquid Ar TPC to search for the proton decay and to observe the solar and cosmic neutrinos, which is called ICARUS (Imaging Cosmic and Rare Underground Signals) project [36].

In 1987, Chen's group at Irvine constructed to a 10 ton liquid Ar chamber and obtained the attenuation length of 3.5 m at the electric field of 0.5 kV/cm using a circulating purification system [37]. In 1988, a Karlsruhe group succeeded in observing three-dimensional images of ionization tracks using a 15 l liquid Ar TPC [38]. In their experiment, they employed a strip type electrode to sense the location of the ionization tracks two-dimensionally. In the last year, the ICARUS group also succeeded in operating a 3 ton liquid Ar TPC with two-dimensional wire electrode system and in observing three-dimensional pictures of tracks produced by cosmic rays [39]. One could expect from these successes that in the near future some massive liquid Ar TPCs will be built for physics experiments.

In 1981, Masuda et al. [40] demonstrated that the electron attenuation length of about 1 m was achievable in liquid Xe at the electric field of 2 kV/cm. They purified Xe gas with a closed Ba–Ti getter system. Due to its limited volume, however, the system was not suitable to purify a large amount of gas. Recently, the collaboration team of the Universities of Waseda and Columbia [11] has shown that an electron attenuation length longer than 1 m was achieved in liquid Xe purified by a semi-open purification system which can treat a large amount of gas. At the time of this conference, the observation of three-dimensional tracking has not yet been realized with a liquid Xe TPC, although two-dimensional tracking has been done by Aprile et al. [42]. However, I expect that the realization of three-dimensional tracking in liquid Xe TPC will be made in the very near future.

5.2. Homogenous liquid rare gas calorimeter

It is a well known fact that the energy resolution of a "homogeneous calorimeter" is superior to that of a "sampling calorimeter". In realizing a "homogeneous calorimeter" based on rare gas liquids, the most important question is what type of electrode structure is best for the calorimeter. In the case of a gridded-ionization-chamber-like structure applied to the "ho-

Table 1
The parameters of prototype models of "homogeneous calorimeters" and their energy resolutions

	Japan group (1985): Liquid Ar calorimeter [43]	Russia group (1990): Liquid Xe calorimeter [44]	Russia–Italy group (1990): Liquid Kr calorimeter [45]
Gap [mm]	9	12	20
Thickness of electrode	1.2 mm (G-10) + 2×18 μm Cu	50 μm Al	0.5 mm (G-10) + 2×35 μm Cu
Energy loss at electrode [%]	18.6	0.48	3.68
Length of calorimeter [cm]	196.5 (13.8 X_0)	50 (17.7 X_0)	76 (16.4 X_0)
Diameter of calorimeter [cm]	102 (7.2 X_0)	25 (9.0 X_0)	42 (9.5 X_0)
Number of readouts	20	1	19
Energy resolution [%]	2.4 (for 1 GeV)	$3.4\%\sqrt{E}$ [GeV]	1.7 (for 1.2 GeV)

mogeneous calorimeter", the distance between the cathode and the grid should be long and as a result, the use of very high applied voltage is required. Alternatively, one could use a multi-parallel-plate chamber in which each plate is so thin that we can approximately consider it to be a "homogeneous calorimeter". In this case, there is no requirement for applying high voltage, and so easing the realization of this type homogeneous calorimeter. Recently, liquid Ar [43], Kr [44] and Xe [45] electromagnetic (e.m.) shower calorimeters of this type were constructed and tested. Table 1 shows the main parameters and their energy resolutions. As can be seen from the table, the energy resolutions reported in this type of calorimeter are much better than those of sampling calorimeters. A 35 ton liquid krypton "homogeneous calorimeter" is now being constructed at the Institute for Nuclear Physics at Novosibirsk for elementary particle physics experiments [46] and is expected to be operated with excellent performance. I would expect that this type of calorimeter will be realized in the near future with an energy resolution better than 1% (rms) for electrons or γ-rays in the GeV region.

5.3. Scintillating liquid rare gas detectors

In 1988, Chen [47] at MIT proposed to construct a liquid Xe scintillating calorimeter as an EM shower detector for the SSC experiment, because of its high photon yield, its fast time response and its adjustable e/π ratio which can be done by choosing and appropriate pulse shaping time. Although this proposal was not adopted in the SSC project, the excellent properties of this calorimeter, compared to liquid ionization calorimeters, still remain intact. Ypsilantis [48] independently proposed the simultaneous use of both signals of ionization and scintillation in e.m. shower calorimeters. The tests of these new calorimeters are done using prototypes. These new ideas are relying upon the recent experiments carried out by Japanese groups on scintillations in rare gas liquids and the relation between scintillation and ionization in liquid Ar obtained during the period from 1978 to 1987.

From 1978 up to 1983, the Rikkyo [49] and Waseda–Riken [50] groups investigated the decay curves of scintillation signals excited by 1 MeV electrons, α-particles and fission fragments in liquid Ar, Kr and Xe. In addition, the Rikkyo group observed the change in the decay curve of scintillation signals of liquid Ar by doping Xe, N_2 and CO_2 [51].

From 1985 to 1989, the Waseda group investigated the relation between the ionization and the scintillation yields produced by relativistic heavy ions in liquid Ar [52–54]. Combining all of these results, they obtained the LET (linear energy transfer $\overline{dE/dx}$) dependence of scintillation yields over a wide range of LET [55]. They also found that the sum of ionization yield and scintillation yield is nearly constant over the wide range of LET [55], and that the energy resolution of the sum signals is almost consistent with the theoretical limit [53]. This result provided a strong motivation to use both signals in e.m. shower calorimeters [48]. Using these data, the Waseda group estimated the W_{Ph}-value in liquid Ar and Xe, which is defined to be the average energy required to produce a scintillation photon in liquid, for relativistic heavy ions, α-particles and 1 MeV electrons [56]. Table 2 shows the estimated W_{Ph}-values in liquid Ar and Xe as well as the value in liquid Xe for α-particles recently reported by Miyajima et al. [57], which roughly agrees with the Waseda value [#1].

Table 2
W_{Ph} values in liquid Ar and Xe for various particles [28]

	W_{Ph} values [eV]		
	Relativistic heavy ions (Ne ~ La)	α-particles	1 MeV electrons
liquid Ar	19.5 ± 2.0	27.5 ± 2.8	25.1 ± 2.5
liquid Xe	14.7 ± 1.5	19.6 ± 2.0 (16.3 ± 0.3) [a]	23.7 ± 2.4

[a] Miyajima et al. [57].

As can be seen from the table, the W_{Ph}-values in liquid Ar and Xe are almost comparable to that in the NaI(Tl) crystal. Even if the light collection efficiency in a liquid scintillation detector is small we can therefore expect a high energy resolution for high energy particles depositing energies larger than 1 GeV. The collaboration team of the MIT, the Universities of Waseda and Columbia and RIKEN [58] observed energy spectra of scintillation produced by relativistic Al ions each of which gives the deposited energy of 2.47 GeV in liquid Xe. As expected, the energy resolutions of ~ 0.5% (rms) were obtained even with the amplifier having a gate width of 40 ns.

To realize such good energy resolution in e.m. calorimeters, good uniformity of reflecting walls, a long attenuation length for scintillation photons, and a high light-collection efficiency are required. Since the R&D program on these issues from the MIT-group can be found in the invited talk given by Chen [59] in this conference, I would like to show our recent result on the attenuation length for photons in liquid Xe. Until quite recently, it was very difficult to obtain an attenuation length for photons longer than ~ 40 cm in liquid Xe even if high purity Xe gas was used. Most recently, Ishida [60] at the Seikei University found that the attenuation length for photons in liquid Xe depends on

[#1] In this conference, Seguinot et al. [48] presented 30–35 eV as the W_{Ph}-value in liquid Xe for 100 keV electrons.

the quality of thermal insulation in the cryogenic system. When the cryostat, without superinsulator for thermal insulation, was evacuated with a rotary pump, the attenuation length attained was ~ 30 cm. When the superinsulator and turbo molecular pump were used, however, an attenuation length longer than 1 m was constantly achieved [#2]. In both experiments, Ishida used the Ba–Ti getter for purification of Xe gas. This result suggests that the light scattering due to the density variation in liquid Xe near the detector and/or the wall limits the attenuation length of photons, and indicates that it is necessary to keep a good thermal insulation during the experiment as well as using high purity gas.

5.4. Photoionization detector

The idea of photoionization liquid detector was firstly proposed by Policarpol [62], University of Coimbra, when he visited Japan in 1980. The real experiment of photoionization detector was made by Anderson at FNAL and Waseda group. In 1984 the Waseda group started the test of a photoionization detector using liquid Ar doped with TMEA. However, since purification of TMEA was very difficult and its solubility in liquid Ar was very small, their investigation was stagnated for several months. In 1986, Anderson reported the result of a systematic survey on relative photoionization efficiencies of many organic dopants in liquid Ar and showed that "allene" (C_3H_4) in liquid Ar has the highest quantum efficiency for photoionization [63]. In his experiment, the liquid Ar and the organic dopants were prepared without purification, so that high quantum efficiency may not have been attained. The Waseda group made measurements of photoionization quantum efficiencies of allene, TMA, TEA and ethylene in liquid Ar and TEA and TMA in liquid Xe. Their results are summarized in table 3. The quantum efficiency of liquid Xe give the highest value of 0.8 among the various combinations of dopants and liquids.

The photoionization effect mentioned above can be used for the following purposes:

1) To improve the energy resolution in low electric field.
2) To improve the sensitivity of ionization chambers for high LET particles.

A typical example of 1) has already been described in section 2. Point 2) is based upon the fact that doping photosensitive organic molecules into liquid Ar or Xe increases the ionization pulse height by the photoionization process of dopant molecules due to recombination photons which are abundantly produced when heavy ions are detected. Recently, the Waseda group has constructed a sampling calorimeter filled with allene doped liquid Ar for energy measurements of relativistic heavy ions and demonstrated an excellent performances for high energy heavy ions [64]. Also, this principle might be used for improvement of the e/π ratio in calorimeter.

Expecting that the devices utilizing rare gas liquids, pure and doped, will be sucessfully applied in various fields of physics experiments in the next decade. I would like to close my review on the history on the R&D of rare gas liquid detectors.

Table 3
Photoionization quantum efficiency ϕ for organic molecules in liquid Ar and Xe

Molecules	Liquid Ar	Liquid Xe
Ethylene	< 0.25 [a]	
Allene	0.6	
Trimethylamine	0.3	0.8
Triethylamine	> 0.2	0.8

[a] Some VUV photons from liquid argon is not absorbed due to high IP of ethylene. The value of ϕ is a lower limit.

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