

Aging in gaseous photodetectors

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Abstract

This paper describes the present status of aging studies in various photosensitive detectors. New experimental data are presented on aging of trimethylamine (TMA) and ethylferrocene (EF) photosensitive vapors as well as on CsI and SbCs photocathodes. A new explanation of the aging process for CsI photocathodes based on solid –state physics theory is given.

Finally, based on our studies, a general conclusion is made that thin polymer deposits on a detector's cathode due to aging can provoke breakdowns through the Malter-type mechanism, or more precisely, via an explosive field emission mechanism.

I. Introduction

An interest in photosensitive gaseous detectors appeared after work was published [1,2] in which the first photosensitive wire chambers were described. These detectors were flushed with benzene or toluene vapors having a relatively low ionization potential. UV photons penetrating through a transparent window caused photoionization and this created primary photoelectrons, which then triggered avalanches. Nowadays, such types

of detectors are widely used in many experiments. The reasons for their success are very simple: they have quantum efficiencies (QE) in the UV region comparable to or even higher than the best vacuum photomultipliers. At the same time, they are position sensitive, cheap, and simple. Of course they are not free from some drawbacks. One of the drawbacks is aging; another is the fact that their spectral sensitivity is limited by the ionization potential. The vapors with the lowest ionization potential E_i used so far are: TMA ($E_i=7.25$ eV) [3], EF ($E_i=6.08$ eV) [4], TEA ($E_i=7.15$ eV) [5], and TMAE ($E_i=5.28$ eV) [6]. Unfortunately, in the last ten years or so, there has been no progress in finding new vapors with E_i lower than EF or TMAE that are compatible with gaseous detectors. This is why an interest arose in pursuing an alternative approach – to use solid photocathodes in gaseous detectors (see [7] and references therein). The threshold of the spectral sensitivity of such detectors is determined by the material work function, which can be very low. However, such detectors could be competitive with those filled with photosensitive vapors only if they are stable in time and if their aging properties are satisfactory.

One should note that there are many more applications of photosensitive materials (vapors and solid photocathodes) than just the detection of UV photons. For example, EF vapors were used a long time ago for plasma studies in so-called 'X-ray cluster counters' in which individual primary electrons produced by X-rays were counted [4,8,9]. This approach was further developed by Breskin's group [10]. TMA, TEA and TMAE are widely used [11] for optical readout of gaseous detectors in so-called 'light chambers' (optical readout [12]). Most of the solid photocathodes are also good secondary electron emitters and can be used in the detection of charged particles and X-ray photons [13].

The successes of all these applications depend very much on good aging properties of the materials used.

In this paper we will review the present status of aging studies in photosensitive detectors. New results on this subject will be presented as well as a new interpretation of aging for CsI and other solid photocathodes.

Finally, a general conclusion will be stated that thin polymer deposits on a detector's cathode due to the aging can provoke breakdowns through the Malter-type mechanism.

II. What is known about aging of photosensitive materials ?

II-1. Photosensitive vapors

Systematic aging studies were done only for wire chambers filled with TMAE and TEA vapors (see Fig.1). It was found that the aging rate depends very much on system cleanliness and anode wire diameter, but only slightly on the composition of the carrier gas.

The analysis of polymer products on the anodes and the cathodes of the wire chambers were done in the past only in the case of TMAE [14]. It was found that on the anode wires the main deposits are not oxidized TMAE, but rather a tertiary amide. It remains unclear however, what fragments of TMAE react with oxygen to form tertiary amide. There have also been findings of some unidentified heavy organic compounds with atomic masses of 202, 110 and 100. On the cathode wires, the following deposits were found: carbon in form of graphite, small fractions of amides (N-C=O), amines NR_3 (R=C,H), and silicone $[\text{R}_2\text{SiO}]_n$ (which probably comes from the oil bubbler). The ratio of elements was as follows: 83% C , 8.2% O , 5.9% N , 2.7% Si .

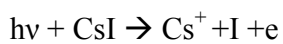
Thus many types of organic species coated the cathode. From a first glance it looks as if at least part of the problem comes from contaminations (for example Si).

The other important consequence of aging was the Malter-type effect, a sporadic burst of electron emission from the cathode. The role of this effect in the detector's operation will be discussed at the end of this paper.

II.2 Solid photocathodes

Various photocathodes were used for various applications. For example, a wire chamber with a CuI photocathode was used successfully for plasma diagnostics [4]. However, the most popular today is the CsI photocathode [15]. There are two main reasons for this: it has high QE (see Fig.2) and allows a short exposure to air, which is very convenient in the process of assembling the detector. Unfortunately, the aging data published so far are very contradictory [16]. This great spread of data is probably due to the fact that aging depends on cleanliness, exposure to air and water vapors, and other poorly controlled parameters.

One should note that the explanation of the CsI aging mechanism is not at all satisfactory. Indeed, the present theory is based on an assumption that CsI is dissociated under UV radiation or ion bombardment (see for example [16]):



This model is certainly correct for CsI molecules. However, in the case of CsI in crystalline form, the aging mechanisms are different. To top it all, in most gaseous detectors operating at 1 atm, the positive ions do not even have enough energy to dissociate the CsI.

III. Experimental set up for aging tests in ultra-clean conditions

The main conclusion, which can be derived from previous studies, is that aging depends on the cleanliness of the experimental set up. In this work we performed aging studies of several photosensitive materials, which were used in some of our applications: TMA, EF, CsI, and SbCs in the cleanest possible conditions.

One should note that it is very difficult (and expensive) to ensure an extremely clean gas system when it is continuously flushed with gas. The main problem comes from the continuous outgassing of the materials used for the construction of the gas system, test chamber, and the detector itself. It is very difficult and takes a long time to outgas the experimental set up by operating in a gas-flushing mode. It is much more efficient to outgas the set up by pumping and heating to a reasonably high temperature and then use the detector in a sealed mode. This is why in this work we chose the latter approach. Our experience shows that if one tries to use the same system (prepared as described above) in a gas flushed mode, then it would be more difficult to ensure the same level of cleanliness as it would be possible with a sealed detector. For example, an additional contamination comes from the oil bubbler, which brings Si-based contamination to the system.

III-1. Set up for EF and TMA aging studies

Our experimental set up for the photosensitive-vapor aging study is presented in Fig.3a. It is basically a station for the production of sealed single-wire counters filled with

photosensitive vapors. It was extensively used in the past for manufacturing sealed detectors for plasma diagnostics. The station includes a single wire counter (SWC), a heating and pumping system, and a gas system. The cylindrical cathode of the SWC (diameter of 15 mm) was made of stainless steel, the anode wires of two different diameters (0.05 mm and 0.2 mm) were made of molybdenum; the anode-cathode dielectric interface was made of glass. The detector had a LiF entrance window with 2 mm diameter. Before filling the gas and sealing the SWC, it was pumped to a vacuum of $\sim 10^{-6}$ Torr and heated to 150°C for several days. The gas system contained two small glass bottles: an empty one and another one filled with a photosensitive liquid (EF or TMA), and a glass cylinder with spectroscopically clean Ar, Kr or Xe (99.999% purity). The EF and TMA were chemically cleaned at the Chemical Lab of the Moscow State University (D. Lemenovsky's lab). In addition, before introducing the liquids into the SWC, they were distilled 6-10 times by "cryo"-transfer liquids from one glass bottle to another and pumped.

III-2. Set up for CsI and SbCs photocathode aging studies

In the past, for the testing of solid photocathodes we used a system that allowed it to perform photocathode aging tests without exposure to air [7]. This was achieved by using manipulators, which allowed a transfer of the photocathodes from an evaporation system to the test chamber. The test chamber was then flushed by a gas at 1 atm. This allowed us to achieve rather good aging characteristics [7]. However, our experience with solid photocathodes shows that the best stability and aging characteristics can be achieved only in a compact sealed chamber. This is because in a system flushed by a gas or in a

continuously pumped system, there is continuous delivery and accumulation of impurities on the photocathode surface. This is why in the present work we performed aging tests in sealed detectors only [17]. The experimental set up for these measurements is presented in Fig.3b. It contains a sealed detector with a solid photocathode, a UV source (Hg lamp) and a reference detector: a photomultiplier or a photodiode monitoring the intensity of the UV radiation. To avoid any aging effect in the reference detector the intensity of the UV flux reaching the reference detector was attenuated by several orders of magnitude.

To separately check contributions to the aging process from the light itself and the action of the light together with positive ion bombardment, we performed measurements in vacuum and the gas atmosphere with small multiplication (factor of 5-10). In addition, some control measurements were done with vacuum and gas-filled Hamamatsu photodiodes: R1187, R1187 (filled with Ar at pressure 1 atm by a technique described in [18]), R414, R250.

III-3 Experimental set-up for the aging studies of CsI secondary electron emitters

As was demonstrated in our earlier work [19], a thin-gap parallel plate chamber with a secondary-electron sensitive layer achieves very good position resolution for X-rays of better than 50 μm in simple counting mode [19]. This approach may find large-scale application in a digital imaging technique [20]. However, the aging properties of such an emitter should be demonstrated. In this work we performed some preliminary aging studies of the CsI converter under the simultaneous action of X-rays and bombardment of positive ions from the gas avalanches. Our set up is shown schematically in Fig.3c. It contains the tested parallel plate avalanche chamber (PPAC) with a stainless steel cathode

coated by the CsI layer, a reference PPAC with a stainless steel cathode but without any CsI coating, and an X-ray gun (6-30 keV). Both PPACs were flushed by a Xe (40%) + Kr(49%) + CO₂(20%) gas mixture at 1atm. The X-ray flux hit the cathodes of both PPACs at shallow angles of about 10⁰. In contrast to our previous studies, in this work we used a uniform (not a porous) CsI layer of 0.4 μm thickness. The coating was done at CERN (Braem Lab). During the transfer to the test chamber, the cathode was exposed to air for a few minutes, but during this exposure the area around the photocathode was continuously flushed by Ar. After installation the test chamber was pumped and heated for 24 hours. Only after that, a working mixture was introduced.

IV. Results

IV-1. Vapors

The results of the aging tests obtained with TMA and EF are presented in Fig.1. For comparison, on the same figure some published data for TEA and TMAE vapors [9] are presented. One can see that the aging properties of EF and TMA are superior. Since TMA and TEA are chemically similar, one can speculate that the difference in aging properties is due to the cleanliness.

IV-2. CsI and SbCs photocathodes

At low accumulated total charges of ~mC/cm² the results obtained with our sealed detectors and Hamamatsu photodiodes were similar: no aging effect was observed at all. Tests with Hamamatsu photodiodes were performed up to a much greater accumulated total charge of 30 mC/cm². The results are presented in Fig. 4 (CsI) and Fig. 5 (SbCs), which show the measured photocurrent as function of irradiation time. Note that some

degradation of the QE observed on the tenth day of continuous light illumination was reversible: after blocking the light for a day the QE returned to its original value. Note also that heating of these photocathodes does not exhibit any QE enhancements. This is in contrast to the photocathodes exposed to air [21].

To our knowledge, the published results on photocathode aging in sealed detectors are very poor (see for example [10,22]) and do not allow any quantitative comparison with our data.

IV.3 Aging of the CsI X-ray converter

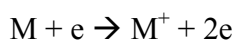
Results of the CsI converter aging test under a combined action of X-rays and avalanches are presented in Fig.6. One can see that no loss of X-ray efficiency was observed up to an accumulated charge of 0.1 mC/cm².

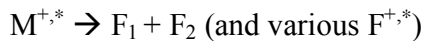
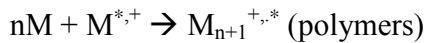
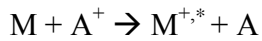
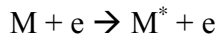
V. Discussions

V-1. Aging of gaseous detectors filled with photosensitive vapors

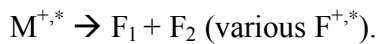
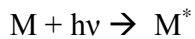
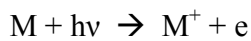
The processes which lead to formation of polymer deposits on the detector electrodes are very complicated. Basically, they include a dissociation of polyatomic molecules M in the gas mixture into fragments F, including ionized (F⁺) and excited (F*) fragments. Schematically these processes can be illustrated as follows.

1) Dissociation of molecules by electron (e) impact, and charge and excitation transfer from the atoms A (atoms of noble gases and atoms appearing due to the dissociation):



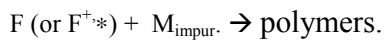


2) Dissociation due to photons (photons of the detected light and photons from avalanches):



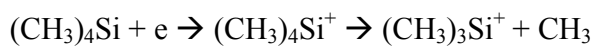
The main result of these processes is the breakage of chemical bonds and the formation of molecular fragments and radicals (bond breaking leads to formation of radicals).

These radicals react either with molecules and fragments or with impurities, for example:



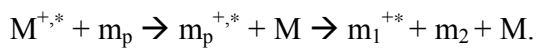
They may also recombine with each other. Thus there are many different channels for polymer formation. Note, that ionized and excited molecules ($M^{+,*}$) are much more chemically active than M. They also have larger cross sections of interactions.

Specific reactions for the photosensitive vapors TMSi and TMP [23], which lead to polymer formation, are:



Even this “very simplified” picture of the processes looks rather complicated.

However, in the case of mixtures with photosensitive vapors Ei_{vapors} is less than $Ei_{\text{main mixture}}$ and this circumstance allows it to simplify the consideration of the processes described above. Indeed, through a multistep exchange processes, all excitation and ionization energy will finally be transferred to the photosensitive molecules m_p (which may then further dissociate into fragments m_1 and m_2):



This was experimentally confirmed by light emission measurements performed for light emission chambers (with optical readout mentioned in the introduction), see Fig.7. As one can see, in mixtures of TEA or TMAE with noble gases or noble gases with mixtures of other molecular gases [24], only emission bands of TEA or TMAE are present in the spectrum. The explanation of this was given in ref. [25]. Thus, in most cases (especially in a low electric field) one should consider only the fragments of the photosensitive vapors in the aging kinematics. This explains why the aging rate depends only slightly on the carrier gas composition, but depends strongly on the nature of the chemical composition of the vapors and on the presence of impurities with which the fragments m_1 and m_2 may react. One can assume that the thickness of the polymer deposits on anode wires depends on the amount of the deposited charge per unit of the anode area.

Consequently, for the same total charge deposited per unit of the anode wire length (i.e. the usual units used for aging results [14]), the aging rate should be approximately inversely proportional to the anode wire diameter, and this was precisely observed experimentally [14].

V-2. Aging of solid photocathodes

V-2a. CsI photocathode

As mentioned above, the explanation of the CsI aging mechanism presented in [10] is not satisfactory at all because it is only applicable to CsI molecules in vapor phase, but not to CsI in solid state phase. In the case of the ideal CsI crystal, the absorption of UV photons should lead to the creation of excitons and free electrons in the conduction band (see Fig. 8a.). This would not cause any dissociation.

However, in reality CsI is not an ideal crystal. It contains cracks, vacancies, interstitials, dislocations, chemical impurities, insertions, and so on. This creates extra energy levels schematically shown as “A-levels” and “B-levels” in Fig. 8b. In this picture A-levels are occupied states above the filled band, and B-levels are unoccupied states below the conduction band. As a result electron traps are formed and new levels appear, from which the photoemission may occur. Note that a CsI photocathode may also may have stoichiometric deviations, i.e. contain excess of Cs or I due to, for example, exposure to light during evaporation or dissociation in water, and this also creates extra levels in the structure of the energy bands.

Dramatic and irreversible changes occur when the CsI photocathode is exposed to air. Its initial polycrystal structure cracks to granules [16]. Absorbed water further damages the photocathode structure and creates a “CsI-water” solution in which Cs and Iodine may dissociate. If an aging test is done with non-monochromatic light, some contribution to the photoemission may come from the created levels and this will enhance the photocurrent. On the other hand, levels can trap free electrons inside the CsI structure and

this may cause a charge-up effect. Thus there are several competing processes, which contribute to the measured photocurrent. If the aging test is interrupted (for example, the light is blocked), some relaxation processes may occur [26] and the degraded QE can be partially restored. The same restoration effect can be expected with photocathode heating. Note that both these predictions based on solid-state theory [26] were confirmed experimentally [21].

Let's now discuss the possibility of CsI dissociation by ion impact (see section II-2). Although the positive ions from the avalanche do not have enough kinetic energy to dissociate the CsI (at least at a pressure of 1 atm and an electric field <10 kV/cm; e.g. in [27]), they can create insertions and with such a process distract an "ideal" crystalline structure. As a result, the photocathode aging characteristics, especially for photocathodes exposed to air, may depend quite extensively on the photocathode history: cleanliness, duration of exposure to air, and so on. This may explain a great range of the experimental data mentioned above.

Thus, the simplified model described in the section II-2 does not at all reflect the complexity of the aging phenomena. One can therefore speculate that aging is a result of a gradual destruction of the ideal crystalline structure through the creation of defects, extra levels, insertions, and dissociation in water layers with a possible further dissociation of Cs and I atoms. Due to the defects, the electron transfer properties will be diminished and the photocathode, layer by layer, will be destroyed, and as a result the QE will drop. This model also explains the heating effect, i.e. the partial restoration of the QE value due to the so-called relaxation effects [26]. All this allows to suggest that the

best aging properties should be exhibited by photocathodes not exposed to air, and our experimental data presented in this work exactly support this prediction.

V-2b. Aging of photocathodes sensitive to visible light

Several groups are now able to manufacture gaseous detectors sensitive to visible light. However, there were earlier observations from Breskin's group [28] suggesting that such photocathodes have very poor aging properties. Our results show that the aging properties of high-quality CsI and SbCs photocathodes (prepared for example by Hamamatsu) are very good, see Figs. 4 and 5. Even when these photocathodes were operated in a gas atmosphere, the ion bombardment did not cause any extra problems. A review of possible mechanisms involved in this type of photocathode aging can be found elsewhere (see for example [22]). In ref. [22] one can also find some discussion concerning the possible correlation between the photocathode resistivity and the variation of the QE with time.

V-2c. Link between the Malter-type effect and breakdowns in gaseous detectors

For simplicity, in the above considerations of photocathode aging, we have neglected the role of deposits on the photocathode surface due to gas aging. However, these deposits may play an important role. We will mention only two major effects here:

- 1) If polymers are formed on the cathode surface, then photoelectrons, created from the cathode, must pass this layer before being able to penetrate to the detector. As a result, some fraction of the electrons will be lost and thus the measured QE will decrease.

2) The deposits (polymer layers or even absorbed layers of gases) can lead to a Malter-type effect - a sporadic emission of electrons [29]. One should note that the classical theory of the Malter effect suggests single-electron emission [29]. In contrast, it was recently discovered that this emission of electrons might occur in the form of bursts or jets of electrons [30]. This effect is very similar to the well-known “explosive” field emission mechanism in vacuum breakdown [31]. Each burst may contain between a few and up to 10^5 electrons emitted in a time interval from a fraction of a μs to ms [29,32]. This Malter-type effect, or more precisely, explosive field emission in turn may lead to two other effects:

- a) a reduction in the photocathode work function (as well as an increase of electron emission from the photocathode), and therefore an increase in its measured QE;
- b) since these electrons receive full multiplication in gaseous detectors they may cause breakdowns [29,32].

This discussion suggests that the gas mixtures in which the photodetectors operate should be carefully chosen: they should not make any deposits either due to polymerization or any absorbed layer. Note however, that in practice, even in well-selected gas mixtures, it is quite impossible to avoid microscopic dielectric insertions on the cathode surface [31]. This may lead to “explosive” field emission. Thus this type of breakdown-triggering mechanism appears to be a general one [32].

VI. Conclusions

Photosensitive materials may have a lot of applications in various fields, but the success of these applications depends on their aging characteristics. This is why studies of their aging behavior are very important.

As a result of our work we can suggest that one of the most important factors in the aging of photosensitive vapors is the system cleanliness.

In the case of solid photocathodes we found that high-quality photocathodes, not exposed to air, have excellent aging properties both in vacuum and in gas atmospheres. This in turn suggests that ion bombardment does not play an important role in the aging of the photocathodes.

However, polymer deposits and even layers adsorbed when exposed to positive ion bombardment may cause QE degradation and a Malter-type effect.

We suggest that this Malter-type effect (or “explosive” field emission) could be the main breakdown-triggering mechanism in any gaseous detectors. This is because in practice it is almost impossible to avoid microscopic dielectric insertions or adsorbed layers on cathode surfaces.

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Figure captions

Fig.1 Results of aging tests of several photosensitive vapors: TMAE (open diamonds) in a not very clean gas chamber [14]; TMAE (open squares and triangles) in a clean gas chamber [14]; TEA (open circles) [14]; EF (filled squares) and TMA (filled circles) - our data obtained with a sealed SWC (anode diameter of 0.05 mm) at gain of $\sim 10^3$; EF and TMA (filled triangles) - our data obtained with a sealed SWC (anode diameter of 0.2mm) operating in streamer mode (gain of $\sim 10^8$).

Fig. 2 Quantum efficiency of several photosensitive materials.

Fig. 3a Experimental set up for manufacturing sealed single-wire counters.

Fig. 3b Experimental set up for aging studies of photocathodes.

Fig. 3c Experimental set up for aging studies of solid X-ray converters.

Fig. 4 Results of aging studies for CsI photocathodes (photocurrent vs. time) manufactured by Hamamatsu: R1187 (open diamonds) and R1187 filled with Ar (filled squares). Note that there were breaks in the measurements between days 5 and 10, 15 and 20, 22 and 25, 26 and 27. One can see that during the breaks the QE value was partially restored.

Fig. 5. Results of aging studies of the SbCs photocathodes manufactured by Hamamatsu: R414 (open diamonds) and R250 filled with Ar (filled squares). Note that there were breaks in measurements between days 5 and 10, 15 and 20, 22 and 25, 26 and 27. One can see that during the breaks the QE value was partially restored.

Fig.6 Ratio of two currents (current from the PPAC with the CsI converter over the current from the PPAC without any converter) vs.time. During these measurements the current value from the PPAC without the CsI converter was about $0.1 \mu\text{A}$ with good accuracy. After 8 hours the X-ray flux was blocked for half an hour and then the exposure to the X-rays was continued.

Fig. 7 Emission spectra of various gas mixtures filled with TEA vapors at total pressure of 1 atm: 1) Ar + 6%TEA, 2) Kr + 6%TEA,3) Xe + 6%TEA.

Fig.8 Energy band scheme for an ideal CsI crystal (a) and for CsI containing various defects (b).

