16

Time degeneracy and ageing

16.1 Early observations

A progressive degradation of performance with the appearance of discharges was observed in the early development of MWPCs, enhanced by the need to operate the detectors at high gains due to the limitations of available electronics. A gaseous detector, in which owing either to improper cleaning or to the creation of deposits on electrodes due to molecular dissociations or polymerization on exposures to radiation, a secondary discharge mechanism is activated, has a very characteristic behaviour (see also Section 7.9). Normally well behaving at low counting rates, a degraded detector shows an increasing background counting rate (or dark current) when exposed even for a short time to higher radiation fluxes; this current may persist after removal of the source.

The appearance and degree of radiation damage can be quantitatively assessed by monitoring the single counting rates at increasing intensities of a radioactive source, as a function of the voltage. Figure 16.1 is a measurement realized by exposing a detector to a ^{55}Fe X-ray source before and after a long-term irradiation of around 10^7 counts/cm^2, operating the counter with an argon-isobutane-freon gas mixture (Charpak *et al.*, 1972). Before irradiation, the efficiency plateau is reached at the same voltage, independently of the rate; the onset of discharges at lower voltages for increasing source intensities is due to spontaneous electron field emission from the cathodes. After a long-term exposure, however, a decrease of the discharge point and a shift to higher voltages of the beginning of the plateau are observed, attributed to an increase of the effective anode wire diameter due to conductive deposits; while hydrocarbon polymers are indeed generally insulating, the anodic layer may be at least partly reduced to elemental carbon by chemical or thermal effects due to energetic electron bombardment in the avalanches.

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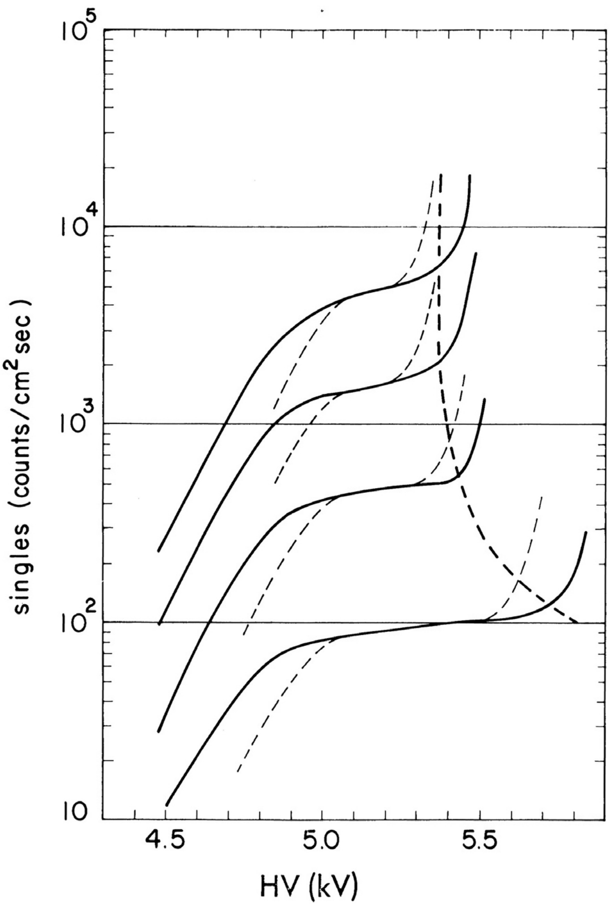

Figure 16.1 Singles counting rates at increasing X-ray fluxes before (full lines) and after a long-term irradiation (dashed lines). 'Magic gas' filling (Charpak *et al.*, 1972). By kind permission of Elsevier.

The use of non-polymerizing additives having an ionization potential lower than any other constituent in the gas mixture, by a process of charge transfer between species (see Section 4.4), was found to prevent the degradation, or at least to increase by several orders of magnitude the integral flux capability of a proportional chamber (Charpak *et al.*, 1972). In Figure 16.2, the singles counting rate plateaux for increasing X-ray source intensities are measured in a chamber operating with 4% methylal[1] added to magic gas; no change is observed in the main operational parameters. Total exposures up to 10^{12} particles/cm^2 have been reported, without detectable ageing effects.

[1] $(OCH_3)CH_2$

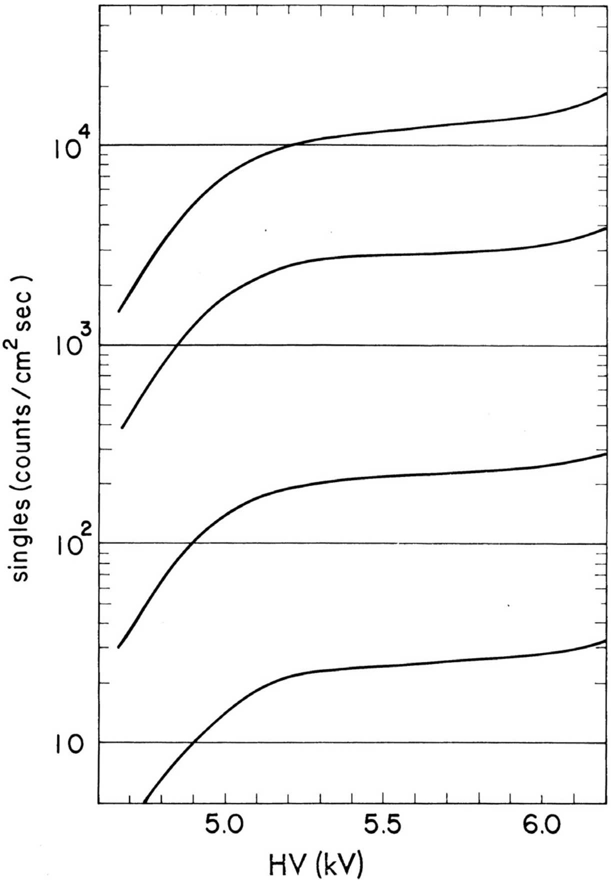

Figure 16.2 Singles counting rates at increasing source intensities measured in a MWPC with 4% methylal added to the 'magic gas' (Charpak *et al.*, 1972). By kind permission of Elsevier.

16.2 Phenomenology of the radiation damages

The radiation-induced damages are local and permanent. Discolourations and thin layers of translucent, whitish or dark materials are often uncovered in the exposed regions, particularly well visible on planar electrodes; Figure 16.3 is a close view of the irradiated area on a chamber with aluminium cathodes (Majewski, 1986); the pattern, possibly due in this case to oxidation by ions released in the avalanches, reflects the pattern of the wires. Damaged wires appear instead darker, suggesting a carbon-like deposit.

Cleaning the damaged regions with various solvents has been attempted, in many cases removing the source of the problems, until the detector is irradiated again. A fast and repeated switching off and on of the operational voltage may

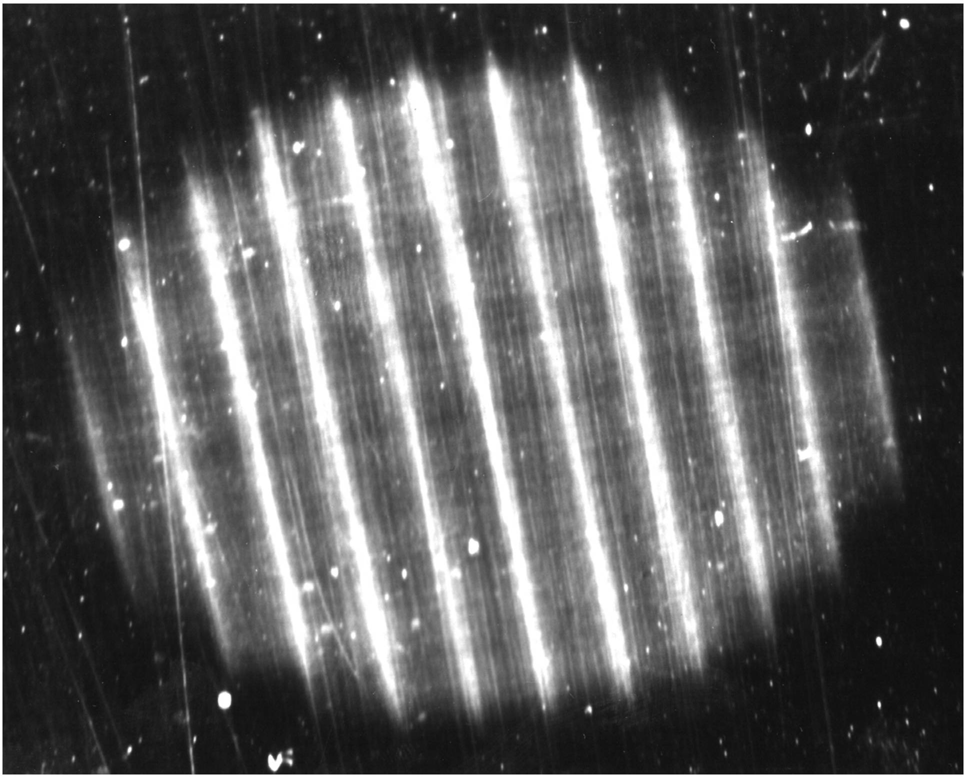

Figure 16.3 Close-up of the damaged area of a cathode after long-term irradiation (Majewski, 1986). By kind permission of LBL.

restore the original low noise operation, probably by capacitive removal of the positive ion deposits on the insulating layers responsible for the secondary sustained discharge, and was actually implemented in early setups to cure the disease, albeit as a temporary solution (Schilly *et al.*, 1970).

The accumulation of positive ions on the thin insulating layers building up on the cathode results in the appearance of a local dipole field, getting stronger the higher the radiation flux; electrons can then be extracted from the layer and injected into the gap by the Malter effect (Malter, 1936). When these electrons reach the anodes they multiply, thus generating more ions; the process easily becomes self-sustaining, and maintains a continuous dark current even after removal of the source of radiation. Temporary suppression of the operating voltage stops the process, which reappears, however, when the detector is exposed to radiation, the more rapidly the higher the flux.

In gas mixtures rich in heavy hydrocarbon quenchers, often used to allow large gains, the conjecture that the degradation is due to a polymerization process

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occurring in the gas under avalanche conditions comes naturally. These polymers, appearing in the liquid or solid phase, deposit on all electrodes, inducing the described secondary phenomena. A similar phenomenology observed in new chambers can be associated with the presence of greasy layers on the electrodes due to improper cleaning.

An obvious way to prevent ageing is to use a gas not containing hydrocarbon additives. However, good energy resolutions and large gains can only be obtained in mixtures having a high photon-absorption cross section, thus preventing secondary effects due to photoelectric emission at the cathode (photons are copiously emitted in the avalanches); this explains the common use of hydrocarbons quenchers. Moreover, it appears that the use of non-polymerizing quenchers, such as carbon dioxide, does not always prevent the ageing process; chemical analysis of the deposits found on damaged wires shows the presence, sometime as dominant species, of plasticizers, silicone and sulphur compounds that could not have been present in the mixture itself but are introduced into the gas flow by the flux regulators, tubes and gas connections.

The standard way to assess the radiation resistance of a detector is to expose it for a long term to an intense radiation from radioactive sources or X-ray generators, and measure at regular intervals the local gain at low rate. Figure 16.4,

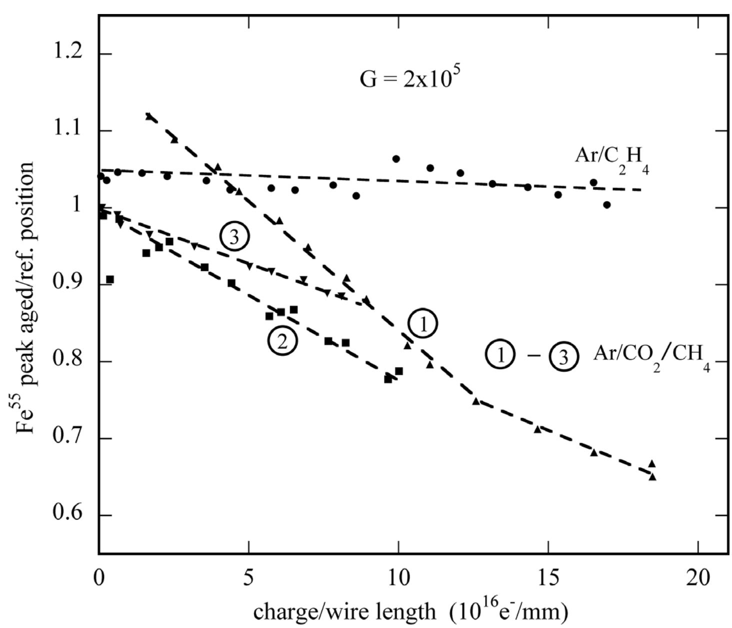

Figure 16.4 Normalized gain as a function of collected charge, measured with a MWPC under sustained irradiation in different gases (Kotthaus, 1986). By kind permission of Elsevier.

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providing the normalized gain as a function of collected charge in several gas mixtures, is a typical example (Kotthaus, 1986). Since the measurement can take weeks if not months, care should be taken to correct for gain variations not related to ageing (temperature, pressure); a convenient procedure is to monitor the detector gain in a non-irradiated part of the chamber.

The effect of gain, materials and gas composition on ageing has been studied for decades; summaries of the status of the knowledge in the field can be found in the proceeding of dedicated workshops and many dedicated articles (Kadyk, 1986; Hohlmann *et al.*, 2002; Capeáns, 2003; Titov, 2004). Aged chambers can be sacrificed to inspect the electrodes, subjecting damaged wires to a wide variety of surface analysis, from microscopy to mass spectrometry. Figure 16.5 collects a representative selection of images of damaged anode wires showing all kinds of deposit, from bulbous layers to crystal-like spikes (Juricic and Kadyk, 1986; Hilke, 1986b). The micron-scale filament growth shown in Figure 16.6 has been found to be mainly composed of silicone polymers, probably seeded by residues of lubricants used in the detector and carried in the gas flow (Binkley *et al.*, 2003).

Despite often contradictory results, from these works emerges a set of general guidelines on how to build and operate gaseous detectors capable of withstanding high radiation fluxes. The 'golden rules' include warnings against the use of heavy organic quenchers, care to avoid sources of silicon oil pollution, indications on good and bad materials for the construction of detectors; detailed tables of 'good' and 'bad' materials are given in Capeáns (2003). Use of additives in the gas flow appears also capable, if not of totally preventing the ageing processes, at least of slowing them down to an acceptable pace (Va'vra, 1986b; Sauli, 2003).

The advent of new generations of detectors, very performing but vulnerable to radiation damage, coupled to the commissioning of higher luminosity accelerators, opened the ageing issue again in all its dramatic relevance. Additives often turned out to have undesirable effects on materials, and the recipes for gas and material choice became more and more difficult to enforce, particularly for large area systems, where cost is an important issue.

Abundant data exist on the dissociation and polymerization processes of hydrocarbons under discharge or plasma conditions; the following references are only representative examples (Lindner, 1930; Yeddanapalli, 1942; Hess, 1986). Many properties of plasma chemistry (Yasuda, 2003) are reminding us of similar observations made in proportional counters, such as the larger polymerization rate of silicone as compared to carbon compounds, and the effects of the electrode material and surface quality. Possible connections between plasma chemistry and gas counters ageing have been made (Va'vra, 2003). The progress in the understanding of the basic underlying chemical and physical processes is often counterbalanced by reports on the dramatic loss of detector systems due to unforeseen

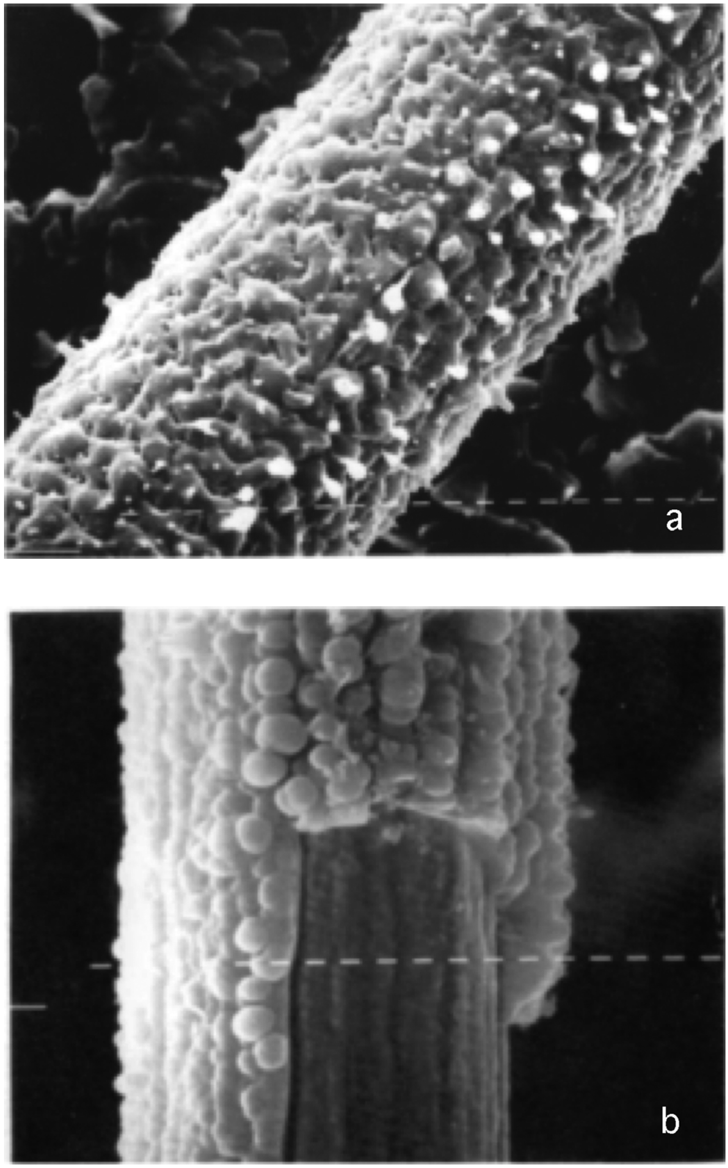

Figure 16.5 Examples of WWPC wires variously coated after irradiation. a: (Ullaland, 1986); b: (Hilke, 1986b); c and d: (Juricic and Kadyk, 1986). By kind permission of LBL.

happenings. Usually obtained in rather specific pressure and charge density conditions, they remain qualitative when extrapolated to the different situations met in gas detectors. Results of systematic investigations on polymer formation in conditions close to those of gas counters have been reported (Kurvinen *et al.*, 2003) and

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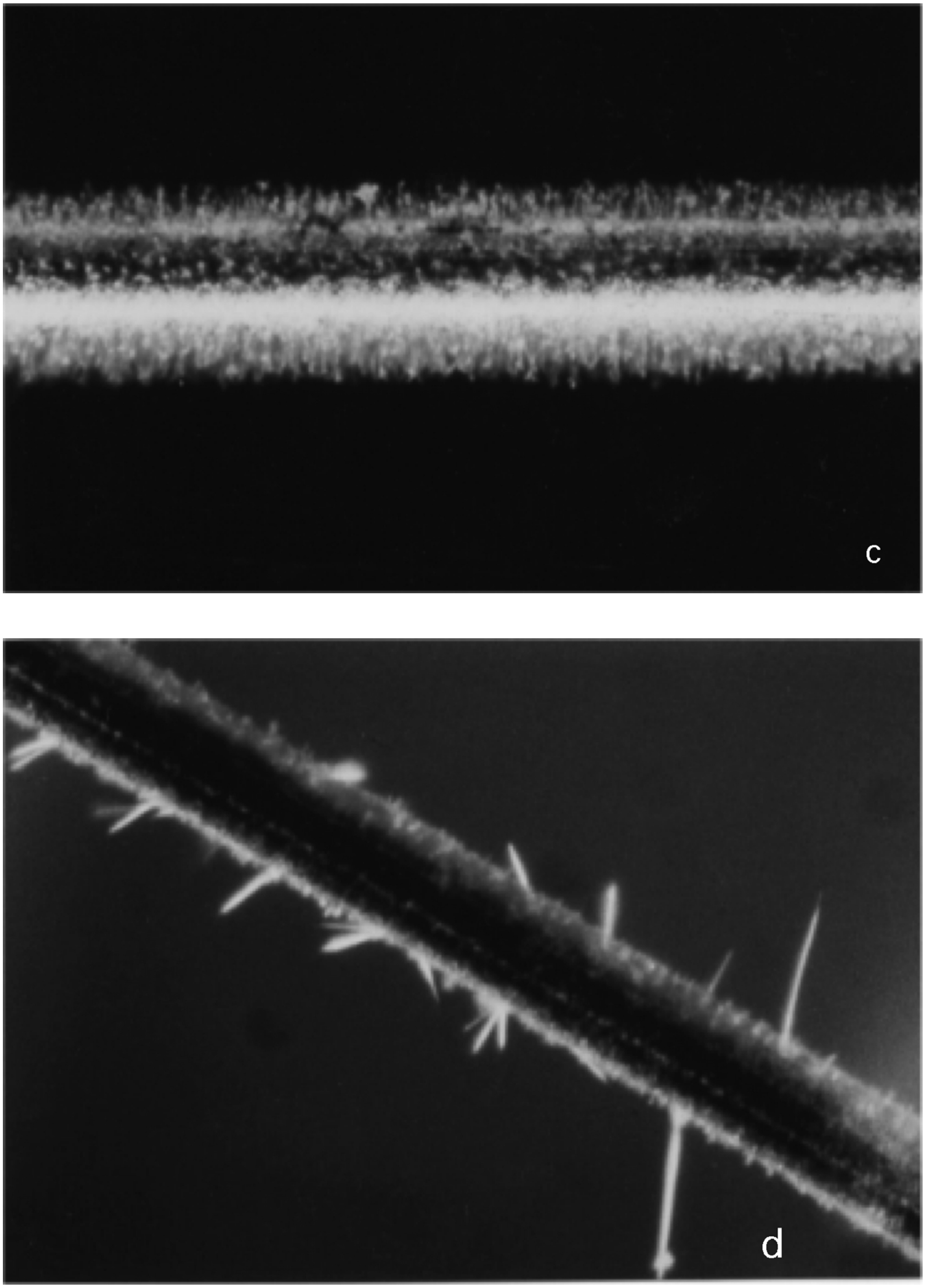

Figure 16.5 (*cont.*)

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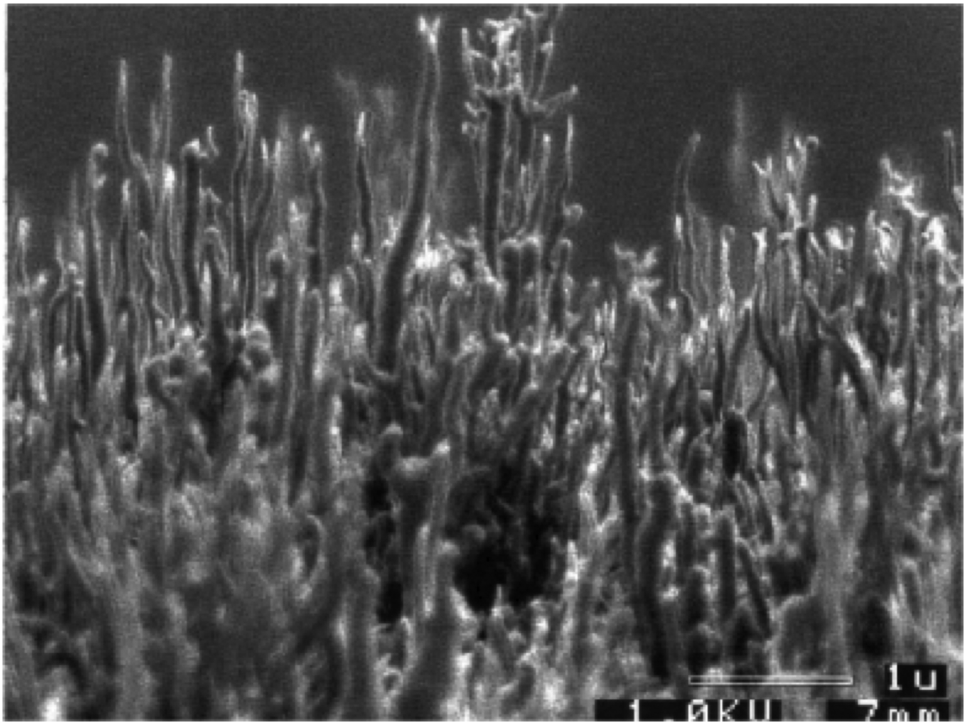

Figure 16.6 Silicone filament growth in a radiation-damaged chamber (Binkley *et al.*, 2003). By kind permission of Elsevier.

confirm the strong reduction of polymer formation with increasing current density, an effect casting doubt on the validity of results obtained with very high rate exposures.

16.3 Quantitative assessment of the ageing rates

The verification in reasonable time of the survival of a detector for several years of operation needs an accelerated test procedure. Intuitively, if ageing is a consequence of the formation and accumulation of polymers or other chemical processes occurring in the avalanches, the scale invariant should be the total accumulated charge, independently of other factors such as voltage, type and flux of radiation, and gas flow. It has become customary to express the accumulated charge in Coulombs per unit length of wire, a detector being qualified good if it could withstand without gain drops up to several C/cm. A quality factor, the ageing rate *R*, defined as the percentage gain variation normalized to the total collected charge, should ideally be small and constant for a given detector (Juricic and Kadyk, 1986). However, it appears that the value of *R* depends, sometimes critically,

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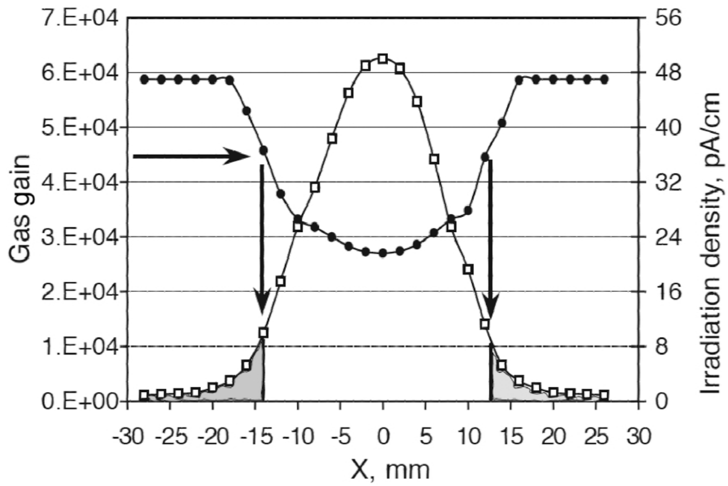

Figure 16.7 Irradiation current density (open squares) and gain reduction (full dots) as a function of position along the wire. Dashed areas correspond to regions of limited streamer formation (Ferguson *et al.*, 2003b). By kind permission of Elsevier.

on test conditions, the main factor being the dose rate, or acceleration factor: data collected at high current densities tend to be optimistic, often by a large factor, compared to those obtained at lower rates (Bouclier *et al.*, 1994b).

Depending on geometry and irradiation conditions, a space charge gain reduction can set in at high current densities, possibly reducing the polymerization efficiency; a reduction of gain by a factor 2 has been measured in the centre of the irradiated area for the measurement shown in Figure 16.7 (Ferguson *et al.*, 2003b). At high operating voltage, the gain on the edges can be large enough to set in a limited streamer regime, thus substantially increasing the collected charge and giving rise to characteristic ring patterns in the damaged irradiated areas.

The appearance of diffused micro-discharges, which produce irreversible local damages or filament growth ('hairs'), can seriously affect performances. Extreme sensitivity to some pollution sources, particularly if containing traces of silicone compounds, has been observed (Ketzer *et al.*, 2001). The interpretation of irradiation results can be seriously biased if these sources, often external to the chamber, are undetected, making comparisons with other observations doubtful.

A correlation between the wire diameter and the rate of ageing has been also found, thinner wires ageing much faster (see for example Va'vra, 1997); this might be simply due to the smaller area available for the deposits, or to an increased polymerization efficiency due to the higher field close to the anode.

Given the dose rate, a larger irradiated area can also increase the ageing rate (Hott, 2003; Hildebrandt, 2003); the gas flow itself affects the ageing rate, larger

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for lower flows. Moreover, the gain degradation appears to move along the detectors with the gas flow, the damaged area often largely exceeding the irradiated region (Hohlmann *et al.*, 2002; Titov, 2004). A possible explanation of these effects lies in a dependence of the polymerization rate on the time that reactive radicals produced in the avalanches stay in the region of ionization before being carried away by the gas flow.

16.4 Methods of preventing or slowing down the ageing process

As discussed in the introduction, the addition of low ionization potential vapours like methylal, ethyl or iso-propyl alcohols to the main gas mixture considerably slows down the ageing process, aside from improving the performances of the detectors thanks to their added photon-quenching properties (Charpak *et al.*, 1972; Atac, 1987).

The beneficial effect of adding water to detectors has also been known for a long time. Aside from reducing the polymerization rates in plasma discharges, water has also the property of making all surfaces in the detectors slightly more conductive, thus reducing, if not the formation of polymers, at least the accumulation of ions on the thin layers, responsible for the gain degradation and the increase of dark current through the Malter effect.

Results of measurements with water addition to the gas mixture are, however, contradictory, as seen by comparing Figure 16.8 (DeWulf, 1986) and Figure 16.9 (Kotthaus, 1986); while in the first case the addition of a few parts per mille of water substantially reduces the ageing rate, in the second case no effect is observed. This is a representative example of outcomes in a field where the presence of trace impurities may dominate the ageing process. In some cases, the culprit has been identified; Figure 16.10 shows the effect on ageing of the addition in the input gas line of a short section of plastic tubing: the gain loss rate in the aged position is increased by a factor of two.

Extensively used in the early experimental setups, the addition of vapours, that requires the use of temperature-controlled bubblers, is not very practical in large systems, and has encouraged the search for alternative methods of control.

Over the years, experience has shown that even a supposedly robust detector can suffer from severe ageing problems due to the unexpected presence of pollutants. A therapy to cure the disease, even exceptionally, would be very welcome. Procedures to evaporate oily deposits on anodes by heating the wires with an externally supplied current were tested, but can only be used with high resistivity wires (Va'vra, 1997). In most cases, however, detectors are not designed to heat up the wires by DC current, either for lack of connections on both sides of the wires of because of their low resistance. An alternative method consisting of operating the

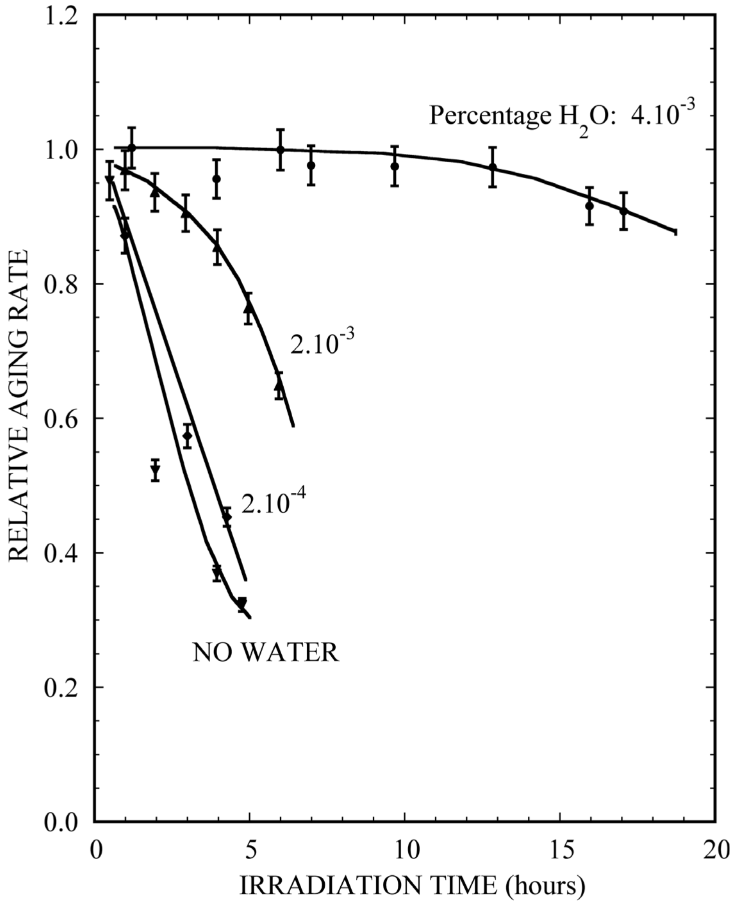

Figure 16.8 An example of the beneficial effect on ageing with the addition of water vapours to the gas mixture (DeWulf, 1986). By kind permission of Elsevier.

detectors in pure argon with a reverse voltage to ‘burn out’ deposits has been tried with some success (Kollefrath *et al.*, 1998).

A rather extreme method of removing the debris deposited on the anodes has been tested by applying a high voltage pulse to ‘zap’ the wires with an energy of a few tens of joules (Marshall, 2003): aged chambers cured using the method recovered and operated successfully. This rather extreme treatment can presumably only be used after removal of the electronics, and it is not clear what the fate of the zapped fragments is; moreover, the sublimation yields can affect the efficiency of the detector.

Operation of a damaged detector with the addition of small amounts of oxygen (∼500 ppm), appears to gradually reduce the current, presumably by burning away the organic deposits (Boyarski, 2003). This method, if applicable in other devices, has the advantage of not requiring special connections to the wires.

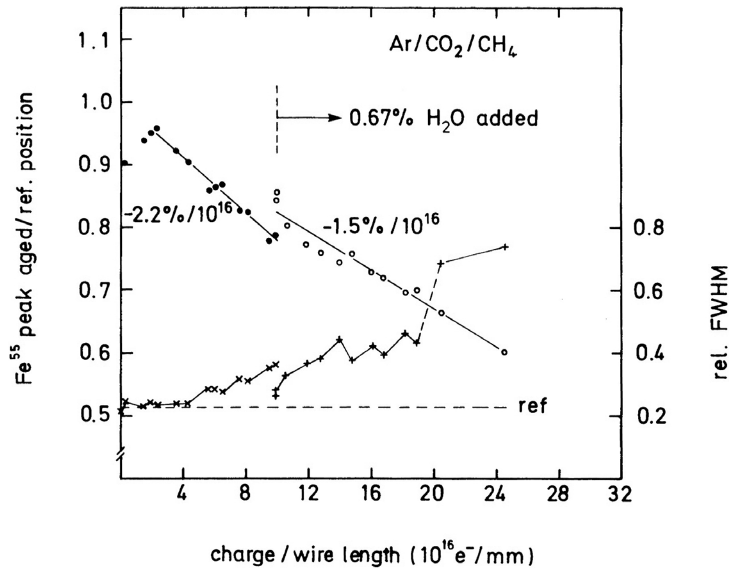

Figure 16.9 In this example, addition of water has no effect on ageing rate (Kotthaus, 1986). By kind permission of Elsevier.

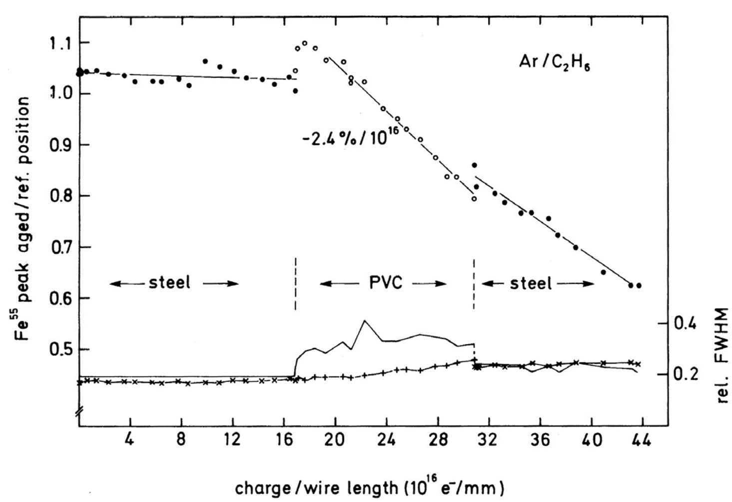

Figure 16.10 Strong ageing under irradiation after introduction in the gas line of a short section of PVC. The degradation continues even after removal of the plastic tube (Kotthaus, 1986). By kind permission of Elsevier.

The presence of silicone deposits on wires, observed when removing all possible sources of organic contamination, has suggested the addition of carbon tetrafluoride to the gas mixture: CF_4 is indeed used in the semiconductor industry to etch silicon in wafer manufacturing. The use of carbon tetrafluoride as a

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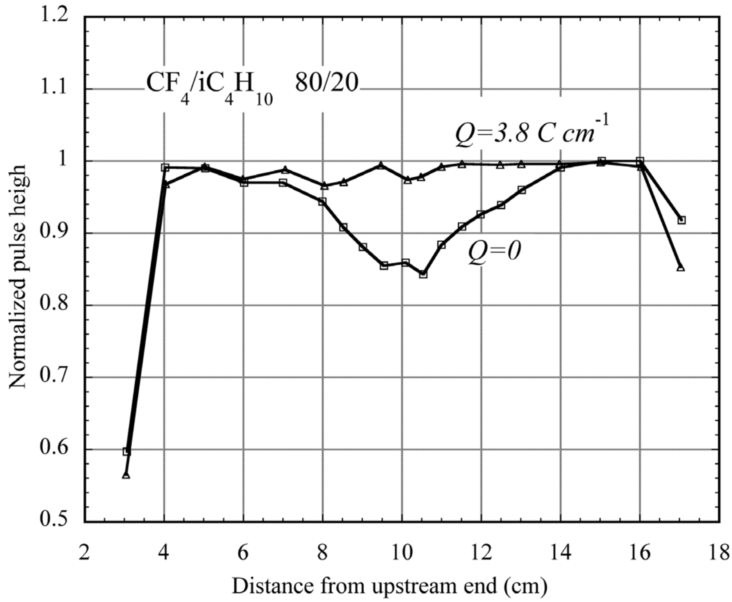

Figure 16.11 Gain scan along a wire of a damaged chamber before ($Q = 0$) and after irradiation with CF_4 in the gas mixture ($Q = 3.8$ C cm^{-1}) (Openshaw *et al.*, 1991). By kind permission of Elsevier.

quencher in gas mixtures is also attractive because of some properties relevant for particle detectors: fast electron drift velocity at moderate fields, non-flammability and low neutron cross section. Non-organic, it does not form polymers, and actually it has been demonstrated that, thanks to the reactivity of the species produced in the avalanches, CF_4 can prevent polymer formation and even remove them from electrodes if already present, as shown in Figure 16.11 (Openshaw *et al.*, 1991), and confirmed by later works (Belostotski *et al.*, 2008). However, this requires a delicate balance between etching and deposition processes, not always easy to achieve in detectors (Capeáns, 2003).

The reactivity and lifetime of species produced by CF_4 in the avalanches, particularly in the presence of organic pollutants, water or oxygen, have been extensively analysed (Schreiner, 2001). It was also found that, when exposing detectors containing CF_4 in the gas mixture, long-lived electro-negative molecules form and propagate with the gas flow (Capeáns *et al.*, 1993); this propagation has been confirmed by other studies (Danilov *et al.*, 2003; Albrecht *et al.*, 2003; Akesson *et al.*, 2002) and can affect the efficiency of large systems having several detectors in series. The design of a re-circulating gas system, required in large experiments, particularly if making use of xenon, is singularly difficult in the presence of species that can react with delicate components such as purifiers and filters.

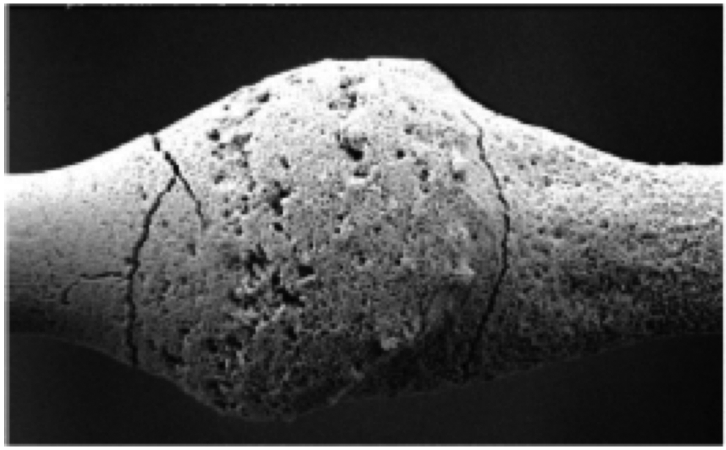

Figure 16.12 Anode wire swelling under the action of fluorinated compounds released by the use of CF_4 in the gas mixture (Akesson *et al.*, 2002). By kind permission of Elsevier.

Damage during operation in CF_4 of the detector's materials, and in particular on the anode wires, has been reported (Akesson *et al.*, 2002; Ferguson *et al.*, 2003a). Under irradiation, presumably because of the penetration of fluorinated compounds into cracks of the wire, the gold-plated tungsten anodes swell and shed metallic flakes; the ill effect of such a happening need not be discussed. The swelling under irradiation has been correlated to the quality of the anode wire surface and to the amount of residual water in the gas, although on the second point the tolerance limits are not clear. The picture in Figure 16.12 gives an extreme example of serious damage to a gold-plated wire in these conditions (Akesson *et al.*, 2002).

The creation in the avalanches of fluorine, that combining with residual water vapours produces hydrofluoric acid (HF), can also have the direst consequences on the constituents of the detector, in particular glass or glass-containing materials, as observed by the ATLAS TRT group: the short glass beads used to join together two segments of long anode wires were seriously degraded under high-flux operation (Akesson *et al.*, 2004b). The use of CF_4 during long-term data taking has been discontinued, in favour of a less aggressive mixture containing oxygen as mild etching agent; short-term runs with carbon tetrafluoride are used to clean the system from silicon containing manufacturing residuals (Akesson *et al.*, 2004b).

16.5 Ageing of resistive plate chambers

The long-term behaviour of resistive plate chambers has been studied extensively in view of their large use in particle physics experiments, in some cases following serious degradation of performance. Problems may arise because of imperfections

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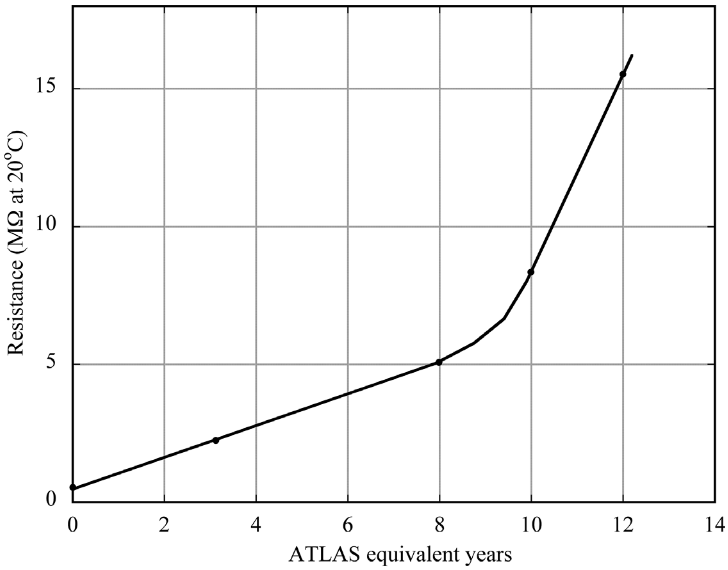

Figure 16.13 Long-term increase of the RPC electrodes' resistivity on exposure to radiation. One Atlas equivalent year corresponds to a particle flux of 100 Hz/cm^2 (Aielli *et al.*, 2002). By kind permission of Elsevier.

in the detector quality, as observed in the first generation of the BaBar RPCs, where the main source of degradation was identified as the incomplete polymerization of the linseed oil used to improve the surface quality of the Bakelite electrodes (Anulli *et al.*, 2003), or could be due to modifications of the electrodes during operation. A common observation is an increase of resistivity of the electrodes, due either to a depletion of the ionic charge carriers or to 'drying out' of the components during operation (Figure 16.13); this results in a decreased rate capability of the detector, from several kHz/cm^2 to a few hundred Hz/cm^2 (Aielli *et al.*, 2002). The intricate chemistry of the linseed oil coating with time and exposure to water has been also studied in detail (Va'vra, 2003). The process can be reversed by adding a small amount of moisture in the gas mixture; a strict control of the ambient temperature and humidity in the experiments is needed to ensure long-term stability of operation (Carboni *et al.*, 2004; Band *et al.*, 2006).

The release of fluorine in the freon-rich gas mixtures used for RPC operation has also been found to be responsible for long-term degradation of glass RPCs exposed to radiation, due to the etching action of hydrofluoric acid on the electrodes; the degradation is particularly fast for detectors operated in the streamer mode.

Observed initially on glass RPCs, this type of chemical damage has been also reported for phenolic laminate-based detectors (Band *et al.*, 2008; Kim *et al.*, 2009).

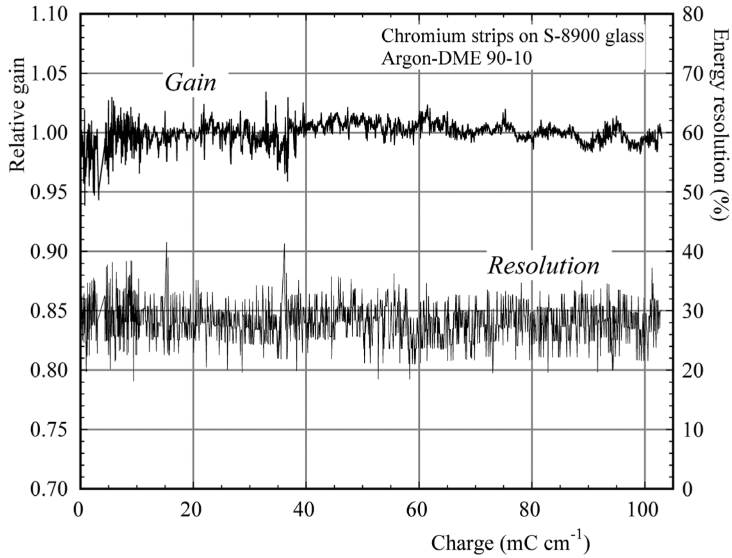

Figure 16.14 Relative gain and energy resolution on 5.9 keV X-rays of a MSGC under long-term irradiation (Bouclier *et al.*, 1995b). By kind permission of Elsevier.

Partial recovery has been achieved by purging the damaged detectors with ammonia-saturated argon (Sakai *et al.*, 2002; Bhide *et al.*, 2006).

16.6 Micro-pattern detectors

Micro-strip gas counters (MSGC) are particularly prone to fast ageing under irradiation, probably because of the small area of the anodes where deposits can easily build up; their radiation tolerance has been extensively studied, in view of the use at high rates and in harsh environments.

While the time evolution of gain is relatively easy to assess with long-term exposures to radiation, the extreme sensitivity of the results to trace pollutants demands a continuous monitoring of the operating conditions and gas purity. In a setup operated at CERN in the framework of an international collaboration to study ageing processes,[2] the gas quality was continuously monitored with a mass chromatograph equipped with a mass spectrometer or an electron capture head to cover a wide range of sensitivity (Bouclier *et al.*, 1996a).

The main results of the measurements, reported in Sauli (1998), indicate that long-term operation of the detector is only possible under strict conditions for the choice of materials and gas purities. Figure 16.14 is an example of long-term gain

[2] RD-28 Collaboration: Development of gas microstrip chambers for radiation detection and tracking at high rates, F. Sauli, spokesman.

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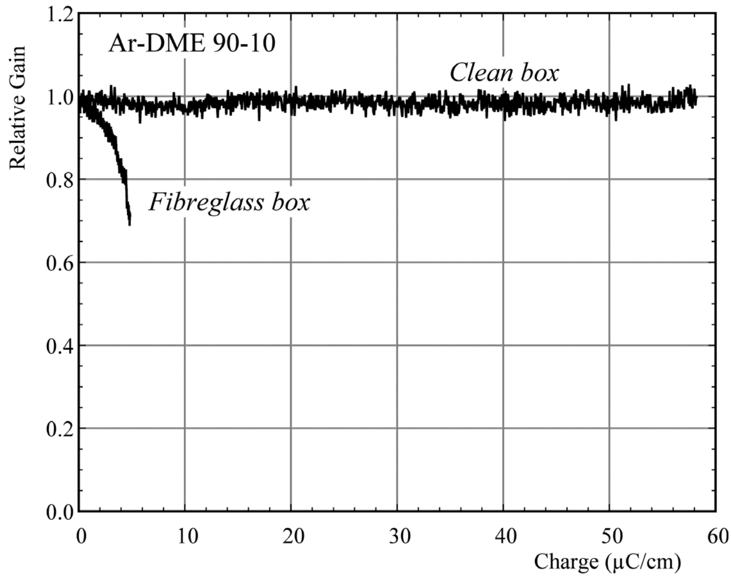

Figure 16.15 Ageing properties of a MSGC plate in a standard and clean assembly (Bouclier *et al.*, 1994a). By kind permission of Elsevier.

and resolution measured with a MSGC operated in argon-DME,[3] under continuous irradiation (Bouclier *et al.*, 1995b); no change is observed up to a total collected charge above 100 mC/cm, corresponding to an integrated minimum ionizing particles flux of about 10^{13} cm^{-2}. It should be noted that, to avoid gain shifts due to insulators charging-up during the irradiation, the measurement can only be performed with detectors manufactured on controlled resistivity substrates, capable of operating at high fluxes (see Section 13.1).

The presence of unwanted pollutants can shorten the detector lifetime by many orders of magnitude. Figure 16.15 is an example, comparing the results obtained with the same type of MSGC plate assembled within a standard fibreglass frame with uncertified epoxies, or mounted in a ‘clean box’, an all-metal and ceramics envelope chemically cleaned and vacuum-pumped before operation; the difference is striking (note the horizontal scale in μC/cm) (Bouclier *et al.*, 1994a).

Results of the systematic search on the effect on ageing of materials and epoxies used for MSGC detectors construction are collected in Bouclier *et al.* (1996a); extended summary tables suggest the recommended choices.

As discussed in Section 13.1, the use of MSGCs has been discouraged by the discharge problems encountered; however, they remain a very sensitive tool to study ageing properties of gaseous detectors.

[3] Dimethyl ether, CH_3OCH_3.

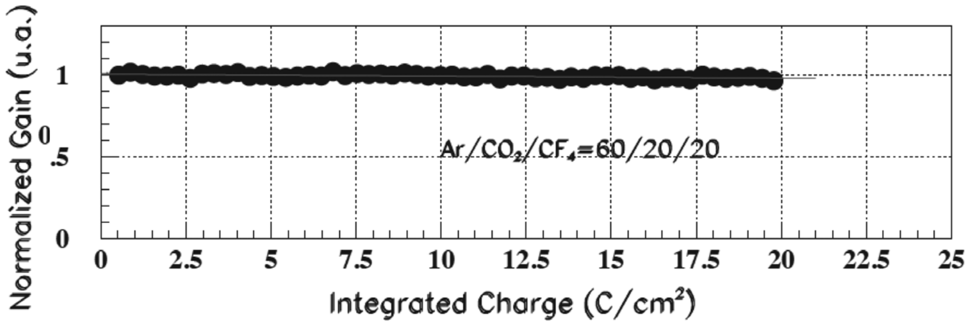

Figure 16.16 Normalized gain as a function of collected charge for a triple-GEM detector (Alfonsi *et al.*, 2004). By kind permission of Elsevier.

The new generation of micro-pattern detectors, Micromegas and GEM in particular, is more tolerant to high radiation fluxes; even though the basic processes of polymer formation in the avalanches are the same as in other counters, their structure does not include thin electrodes, which can be easily coated by deposits with ensuing field modifications. In accelerated ageing tests, realized with continuous exposure to high-rate X-rays, no sign of degradation has been observed with an argon-CO_2 gas filling up to an accumulated charge of 1 C/cm^2 (Guirl *et al.*, 2002; Altunbas *et al.*, 2003).[4]

The addition of carbon tetrafluoride to the mixture, thanks to its etching properties, prevents the most insidious source of ageing due to silicon compound deposits; however, the use of CF_4 in detectors requires special precautions, due to the reactivity of fluorine with water that can generate hydrofluoridric acid, very corrosive for many materials. An example of long-term measurement of normalized gain as a function of accumulated charge for a triple-GEM detector, filled with an Ar-CO_2-CF_4 mixture, is given in Figure 16.16, showing no change up to an accumulated charge of ~20 C cm^{-2}; taking into account the gain of the detector, this corresponds to an integrated flux of about 4×10^{14} minimum ionizing particles per square centimetre (Alfonsi *et al.*, 2004). Similar results have been reported for Micromegas-based detectors (Puill *et al.*, 1999; Kane *et al.*, 2003).

Further reading

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[4] Unlike wire- and microstrip-based detectors, for which the flux given per unit length of the anodes, for devices having continuous electrodes it is expressed per unit surface.