RECOVERY OF GAS DISCHARGE DETECTORS FROM MALTER EFFECT

G.E. Gavrilov, E.V. Kuznetsova, O.E. Maev, D.A. Maysuzenko, S.A. Nasybulin

1. Introduction

Aging or degradation of operating characteristics of gas discharge detectors remains a real problem in modern high energy physics experiments with high radiation background levels. According to results of numerous studies [1, 2], very often aging of gaseous detectors is caused by residual contamination of silicon compounds in the working gas mixture. Silicon is the most widespread element which is regularly registered in analyses of precipitates formed on electrode surfaces. The working gas mixture can be contaminated by a gas circulation pump, gas system connections, and fiberglass electrodes of the detector.

Polymerization reactions take place near the wire in the gas discharge plasma, and the formed polymers precipitate on the surface of anode wires as well as on cathode planes, causing degradation of operation characteristics of the detector. The presence of dielectric polymer films on the conducting metal cathode surface (wires or plane) as a rule results in appearance of a self-sustained secondary emission current, the so-called Malter effect (ME) [3, 4]. This current is the result of accumulation of a positive electric charge on a thin (< 1 μ m) film during the operation of the chamber. Depending on the charge value and the thickness of the dielectric film, the resulting electric field may become sufficient to cause spontaneous secondary electron emission-from the cathode.

For gas discharge detectors, the ME is the most devastating of all secondary electron emission effects. It appears when formation of the electric charge is not compensated by a leakage current from the film surface. Multi-wire proportional chambers (MWPC) operating in a high radiation environment may demonstrate a self-sustained current reaching up to $30-50 \mu A$. The ME results in an increased noise rate, in trips of the high-voltage supply system due to high currents exceeding the usual safe limits, and in the accelerated aging of anode wires around the zone of ME occurrence.

A number of MWPCs operated by the LHCb experiment at the Large Hadron Collider (LHC) demonstrate appearance of the ME. In this work, a noninvasive recovery method of quick and efficient ME current suppression is described. This approach was successfully applied to problematic chambers of the LHCb muon detector and brought them back to a nominal operation. Recovery of gas discharge detectors from aging without disassembling and repair is topical for many modern experiments and often is a challenging task.

2. Curing Malter effect in multi-wire proportional chambers in presence of CF₄

The ME is well known to experimentalists for many years. In some cases it can be cured by adding up of 0.2% of water vapour or 2–3% of various alcohols to the working gas mixture [2, 5]. Molecules of water and alcohol stop the polymerization processes in the gas mixture and precipitation on the cathode surface, and rapidly increase its conductivity. However, these additives cannot be applied to all types of MWPCs. Water vapours deposited on the surface of electrodes and insulators cause micro-sparks, and alcohol vapours, being solvents, cause deformation and swelling of dielectric elements of the detector construction [5].

Because of the above limitations, neither water nor alcohol vapours were considered for eliminating the ME in MWPCs of the muon detector at the LHCb experiment. At the same time, the presence of CF_4 in the working gas mixture of the muon detector (40% Ar + 55% CO_2 + 5% CF_4) allows one to cure the ME without using potentially dangerous admixtures.

 CF_4 is often added to MWPC gas mixtures to prevent chamber aging caused by silicon deposits on anode wires [1, 6]. Moreover, CF_4 is widely used in microelectronics and microelectromechanical device production for plasma etching of Si, SiO₂, and, in presence of oxygen, of various polymer materials [7–11]. Fluorine radicals produced in plasma react with silicon and polymers, resulting in the surface etching by forming volatile products. In proportional chambers, the most intensive formation of free radicals takes place around anode wires, where the electric field reaches 20–200 kV · cm⁻¹. Electron impact dissociation of CF₄ molecules at the electron energy of about $E_e \approx 3-5$ eV occurs with formation of the following chemically active radicals [7, 8]:

$$e^{-} + CF_4 \rightarrow CF_3^+ + F^{\bullet} + 2e^{-}, \tag{1}$$
$$e^{-} + CF_4 \rightarrow {}^{\bullet}CF_2 + F^{\bullet} + e^{-} \tag{2}$$

$$e^{-} + CF_4 \rightarrow {}^{\bullet}CF_3 + F^{\bullet} + e^{-},$$

$$e^{-} + CF_4 \rightarrow {}^{\bullet}CF_2 + 2F^{\bullet} + e^{-}.$$
(2)
(3)

 $^{\circ}$ CF₃, $^{\circ}$ CF₂, and F $^{\circ}$ radicals produced in plasma-chemical reactions (1–3) efficiently react with different silicon formations. Volatile molecules (CO₂, O₂, and SiF₄) formed in etching reactions (4–6) are easily removed from the detector volume with the gas flow:

$$4\mathbf{F}^{\bullet} + \mathbf{S}\mathbf{i} \to \mathbf{S}\mathbf{i}\mathbf{F}_4\uparrow;\tag{4}$$

$$4\mathbf{F}^{\bullet} + \mathrm{SiO}_2 \to \mathrm{SiF}_4 \uparrow + \mathrm{O}_2 \uparrow; \tag{5}$$

$$\mathrm{Si} + {}^{\bullet}\mathrm{CF}_3 + \mathrm{F}^{\bullet} + 2\mathrm{O} \to \mathrm{SiF}_4 \uparrow + \mathrm{CO}_2 \uparrow.$$
(6)

To recover MWPCs from the ME caused by silicon or organic films on the cathode surface, the corresponding depositions should be etched. However, near the cathodes, which are located at a distance of several millimeters from the anode wires, the concentration of fluorine radicals is low. Therefore, the recovery procedure often requires a relatively long time.

3. Recovery procedure developed for the muon detector of the LHCb experiment

The muon detector consists of 1368 modules of 19 different types covering a total area of 435 m². Despite different dimensions, each module has the same internal geometry and consists of four or two MWPC gaps, A, B, C, and D, as shown in Fig. 1. Cathode electrodes of the MWPCs are made of FR-4 fiberglass plates with two-sided 35 µm thick copper coating. Cathode plates of adjacent gaps are separated



Fig. 1. Cross section of the LHCb muon chamber

by honeycomb panels or rigid polyurethane foam which provide precise gap alignment over the whole chamber area. The general parameters of all MWPCs are the same: the anode planes are centered inside 5 mm gas gaps and are formed from 30 μ m diameter gold-plated tungsten wires stretched with 2 mm steps. All MWPCs are filled with the 40% Ar + + 55% CO₂ + 5% CF₄ working gas mixture. The operation gas gain ranges between 4.6 \cdot 10⁴ and 8.8 \cdot 10⁴, depending on the high voltage settings. Each MWPC gap has an independent high-voltage channel. The gas mixture is supplied into the gaps sequentially, as is shown in Fig. 1.

Since the beginning of the LHCb operation, up to 17% of the muon detector modules experienced operation problems caused by

the ME. In average, about 100 MWPC gaps suffer every year from high Malter currents and the concomitant high voltage trips. Nevertheless, all those problematic modules were successfully recovered *in situ*, under the nominal LHC beam conditions by means of a long-term high voltage training with the working gas mixture.

The gaps affected by the ME were trained during data taking. To increase concentration of fluorine radicals, *i. e.* to make the training more efficient, a sufficiently high Malter current should be supported in the damaged zone.

The training session starts with a slightly increased high voltage which provokes a rapid appearance of the Malter current from the problematic region. Usually the initial voltage values do not exceed 20–30 V above the nominal working point. During the training, the high voltage is varied to support the current at the level of ~ 40 μ A.

A MWPC is considered to be restored when the current at the operating voltage reaches the nominal value corresponding to the LHC beam conditions (100-200 nA), and, when the beam drops, the current in the chamber also drops to zero. The duration of the training procedure for restoring a single ME spot in the proportional chamber may vary from one week to several months. A typical MWPC recovery under the high voltage training is demonstrated in Fig. 2. The bottom plot shows a history of changes of the high voltage which supported a reasonable level of the Malter current during the training procedure (top plot). As a result of the training, the self-sustained current dropped down to zero, and the nominal beam current ($I_{nom} \approx 140 \text{ nA}$) was obtained at the working voltage ($V_{\text{nom}} = 2600 \text{ V}$). The total duration of the presented recovery procedure is rather short - five days. On a statistical basis, the results of the training procedure are very positive - more than 90% of the MWPC suffered from the ME were restored to their normal behaviour.



Fig. 2. Typical recovery procedure from the ME at the beam

With all the success of this method, the long duration of the training procedure greatly complicates the possibilities of its application. That is why it was proposed to use an oxygen admixture during the training procedure in order to accelerate the chamber recovery.

4. Gas composition for accelerated recovery from the Malter effect

Various studies of silicon dry etching processes showed that the etching rate in the CF_4-O_2 mixture is significantly higher than that in the pure CF_4 plasma [7–10, 12]. Oxygen radicals promote formation of $^{\circ}COF_x$, which quickly dissociates in collisions with surrounding electrons and atoms (see Eqs. (7–10)), and indirectly increase the number of fluorine radicals in the gas discharge plasma:

$$O^{\bullet} + {}^{\bullet}CF_3 \rightarrow {}^{\bullet}COF_2 + F^{\bullet}; \tag{7}$$

$$O^{\bullet} + {}^{\bullet}CF_2 \rightarrow {}^{\bullet}COF + F^{\bullet}; \tag{8}$$

$$e^{-} + \operatorname{COF}_{2} \to {}^{\bullet}\operatorname{COF} + F^{\bullet} + e^{-}; \tag{9}$$

$$O^{\bullet} + {}^{\bullet}COF \to CO_2 + F^{\bullet}.$$
 (10)

The highest silicon etch rate is obtained in the CF_4 - O_2 mixtures with the oxygen content from 10 to 30% [7–10]. Moreover, oxygen plays a significant role in polymer film dry etching with CF_4 - O_2 mixtures [11] and may even be sufficient in some cases for dielectric deposition cleaning in the pure O_2 plasma [13].

Kinetics of chemical reactions in MWPC gas discharges may significantly differ from reactions rates in industrial reactors due to different plasma nature, gas pressure and electric field configuration. In MWPCs, the electric field varies from $E \approx 5-6$ kV · cm⁻¹ on the cathode surface up to $E \approx 150-200$ kV · cm⁻¹ on the anode wire. In industrial etching set-ups, the electric field is uniform and amounts to 10-50 kV · cm⁻¹, and a silicon wafer (to be etched) is placed in the plasma rich of active radicals. In proportional chambers, radicals are produced close to the anodes, which are separated from the cathodes with several millimeters of a low field region. Moreover, gas-discharge detectors usually operate with gas mixtures at the atmospheric pressure, which is several orders of magnitude higher than the pressure in industrial plasma reactors.

Nevertheless, even though the molecules and radicals in MWPCs have significantly smaller mean free path between electron collisions (~ 1 μ m), the average electron energy (5–10 eV) is quite similar for both proportional chambers and reactors [14]. This makes possible to use dry etching chemical models in qualitative predictions for chemical processes in MWPCs.

Reactions of oxygen impact dissociation and excitation significant at this electron energy range are given below. The oxygen radical production in dissociative electron attachment by O_2 molecules (Eq. (11)) happens already at the electron energies ~ 5 eV, while, for example, the CO₂ electron impact dissociation starts only at ~ 13 eV [15]. Both, the atomic oxygen, O[•], and the excited molecular oxygen *O₂, formed in collisions with electrons (Eq. 12), are chemically aggressive:

$$e^- + O_2 \to O^- + O^{"}, \tag{11}$$

$$e^{-} + O_2 \rightarrow {}^*O_2 + e^{-}.$$
 (12)

The atomic oxygen O^{••} interacts with O₂ molecules forming ozone:

$$O'' + O_2 \to O_3^*. \tag{13}$$

The excited ozone molecule, O_3^* , loses the excess energy through interaction with the plasma. The ground state ozone, O_3 , can participate in processes of plasma chemistry etching on the cathode surface or recombines with free oxygen radicals:

$$O_3 + O^* \to 2O_2. \tag{14}$$

In contrast to the plasma reactor case, the O_2 content in the MWPC working gas mixture should be strongly limited due to the high oxygen electron attachment coefficient. Oxygen reduces the electron density in the discharge plasma, which results in reduction of the charge amplification. Therefore, to keep the gas



Fig. 3. Electron attachment coefficient as a function of the electric field strength (*top*); the gas gain as a function of the oxygen content (*bottom*)

gain at the level sufficient for the recovery process, the oxygen content should be optimized.

To find the optimal amount of oxygen additive in the working gas mixture, simulation studies for muon detector MWPCs were performed using the GARFIELD software package. The results of the simulation are shown in Fig. 3. The electron attachment coefficient as a function of the electric field strength is shown in the top plot. The bottom plot demonstrates the gas gain dependence on the oxygen content in the 40% Ar + 55% CO_2 + + 5% CF₄ gas mixture. Figure 3 (top) shows that for 1-4% of the O_2 content the electron attachment coefficient increases substantially only in the drift region, especially near the cathode surface, where the electric field strength is about $6\,000 \,\text{kV} \cdot \text{cm}^{-1}$. When the oxygen content exceeds 10%, a noticeable electron attachment occurs throughout the whole drift path and in the avalanche region. As a result, the MWPC gas gain at the operation voltage drops by more than 60%, as can be seen in Fig. 3 (bottom). Under such conditions, the charging of a dielectric deposit film on the cathode surface will be slowed

down, which reduces the Malter current. A too small current cannot support an effective etching process. According to the GARFIELD predictions, to compensate for such a gain reduction, the high voltage should

be increased up to 3 000 V, that is above the safe operation range. Therefore, the oxygen content in the gas mixture used for the recovery procedure is a compromise between the acceleration of training and the voltage increase needed to compensate for the gas gain drop caused by the presence of oxygen.

5. Accelerated recovery of muon multi-wire proportional chambers with the gas mixture containing oxygen

To test the ME suppressing procedure with an oxygen-containing gas mixture, four modules of the muon detector were chosen. These modules were removed from the experimental set-up because of high ME currents impeding their proper operation and due to multiple fails of the standard recovery procedure with the nominal working gas mixture.

Localization of the ME zones on cathodes of the damaged MWPC gaps were performed with a collimated ⁹⁰Sr ($E_{\beta} = 2.28 \text{ MeV}$) β source. The Malter currents ignited by the source were hundred times larger than the ionization current. As a result of thorough scans performed for each of the four MWPC of every module, seven ME zones were identified.

The recovery training with an oxygenated gas mixture was performed for each of those zones. The 90 Sr β source irradiated the recovering area to ignite and to support the Malter currents. The training was identical to the one described in Section 3. Supporting a few tens microampere current during the training resulted in the accelerated recovery of all ME zones. The recovery procedure is based on the plasma-chemical etching of silicon and organic compounds by fluoride, oxygen active radicals and ozone produced in the gas discharge. Volatile compounds formed during the etching were removed in the process of gas flushing [4].

Figure 4 shows the currents caused by the ME as a function of the training time for the nominal 40% Ar + 55% CO₂ + 5% CF₄ gas mixture (*red rhombuses*) and for the nominal gas mixture enriched with two percent oxygen (*blue circles*). An initial Malter current of 25 μ A was ignited by the ⁹⁰Sr β source at the voltage of 2 600–2 700 V and was maintained by increasing the voltage in 50 V steps. The ME current decreases as the dielectric film is being removed from the cathode surface. The total time required to recover the ME zone in the case of the oxygen-containing mixture was around 4 h. However, with the nominal working mixture the Malter current stays constant even after more than 6 h [4].



Fig. 4. Current in the MWPC during the ME suppression training

6. Conclusion

- Adding of oxygen to the Ar–CO₂–CF₄ gas mixture allow us to speed up the ME suppression training hundreds of times.
- The given technique allows several times to increase the durability of the MWPC operation in the field of intensive irradiation.
- The non-invasive character of the recovery technique makes it actual for many experiments where the detectors operate with the gas mixtures containing CO₂ and CF₄.

References

- 1. M. Capeans, Nucl. Instrum. Meth. Phys. Res. A 515, 73 (2003).
- 2. S. Belostotski et al., Nucl. Instrum. Meth. Phys. Res. A 591, 353 (2008).
- 3. L. Malter, Phys. Rev. 50, 48 (1936).
- 4. G.E. Gavrilov et al., Phys. Elem. Particles Atom. Nucl. 49, No. 1, 33 (2018).
- 5. J. Va'vra, Nucl. Instrum. Meth. Phys. Res. A 515, 1 (2003).
- 6. G.E. Gavrilov, V.M. Vakhtel et al., Phys. Atom. Nucl. 80, No. 9, 1 (2017).
- 7. J.W. Coburn, Plasma Etching and Reactive Ion Etching, Am. Vacuum Soc., New York, 1982.
- 8. K.R. Ryan, I.C. Plumb, Plasma Chem. Plasma Process. 6, Iss. 3, 205 (1986).
- 9. Yu.N. Grigoryev, A.G. Gorobchuk, Comput. Technol. 8, No. 2, 53 (2003).
- 10. F.I. Grigoryev, *Plasma-Chemical and Ion-Chemical Etching in Microelectronics Technology, Textbook,* Mosc. Gos. Univ., Elektron. Mat., Moscow, 2003 [in Russian].
- 11. M.D. Koretsky, J.A. Reimer, J. Appl. Phys., 72, No. 11, 5081 (1992).
- 12. C.J. Mogab, A.C. Adams, D.L. Flamm, J. Appl. Phys. 49, 3796 (1978).
- 13. Boing Aerospace Research & Engineering Div., Active Cleaning Technique for Removing Contamination from Optical Surface in Space, Final Report D180-17610-1, Contract NAS8-26385, Siettle, Washington, 98124, Aug. 1973.
- 14. J. Va'vra, Nucl. Instrum. Meth. Phys. Res. A 252, 547 (1986).
- 15. J.W. McConkey, C.P. Malone, P.V. Johnson et al., Phys. Rep. 466, 1 (2008).