Characterization of tetrafluoropropene-based gas mixtures for the Resistive Plate Chambers of the ALICE muon spectrometer

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ABSTRACT: The muon identification system of the ALICE experiment at the CERN LHC is based on Resistive Plate Chamber (RPC) detectors. These RPCs are operated in the so-called maxi-avalanche mode with a gas mixture made of tetrafluoroethane (C₂H₂F₄), sulfur hexafluoride (SF₆) and isobutane (i-C₄H₁₀). All of these components are greenhouse gases: in particular, the first gas is already phasing out of production, due to recent European Union regulations, and its cost is expected to increase in the near future. Therefore, finding a new eco-friendly gas mixture has become extremely important in order to reduce the impact of the RPC operation on the environment, and for economic reasons.

Due to the similar chemical structure, hydrofluoroolefins appear appropriate candidates to replace C₂H₂F₄ thanks to their very low GWPs, especially tetrafluoropropene (C₃H₂F₄) with the trade name HFO1234ze(E).

In order to identify an eco-friendly gas mixture fulfilling the requirements for operation in the ALICE environment in the coming years, a dedicated experimental set-up has been built to carry out R&D studies on promising gas mixtures.

Measurements have been performed with a small-size RPC equipped with the front-end electronics, providing signal amplification, developed for ALICE operation at high luminosity after the LHC Long Shutdown 2. HFO1234ze(E)-based mixtures with the addition of CO₂ are discussed in this paper as well as the role of i-C₄H₁₀, as quencher, and SF₆, as strong electronegative gas, in such mixtures.

KEYWORDS: Gaseous detectors; Resistive-plate chambers

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1 Introduction

ALICE (A Large Ion Collider Experiment [1]) is a general-purpose heavy-ion experiment at the Large Hadron Collider (LHC), studying ultra-relativistic heavy ion collisions to detect and characterize the Quark-Gluon Plasma (QGP) [2].

Heavy-flavored hadrons (containing charm or beauty quarks) and quarkonia ($c\bar{c}$ and $b\bar{b}$ bound states) are sensitive probes of the medium created in ultra-relativistic heavy ion collisions [3–5]. Heavy-flavors and quarkonia are studied via their muonic decay channels with the ALICE muon spectrometer [1, 6] at forward rapidity ($2.5 < y < 4$). The spectrometer consists of a set of absorbers, a large dipole magnet, a tracking system made of ten planes of cathode pad chambers arranged in five stations, and a muon identification system.

The muon identification system [7] of the experiment is made of 72 single-gap Resistive Plate Chambers (RPCs [8]). The system has two stations, located at a distance of 16 m and 17 m from the interaction point and arranged perpendicularly around the beam pipe/shielding. Each station consists of two planes, each composed of 18 RPC detectors, covering a total area of about $5.5 \times 6.5$ m$^2$ per plane. The RPC detectors have electrodes (about $270 \times 70$ cm$^2$, 2 mm thick) in phenolic bakelite with a resistivity of $10^9 \div 10^{10}$ Ω cm [9] separated by a gas gap of 2 mm, and the gas mixture composition is 89.7% C$_2$H$_2$F$_4$, 10.0% $i$-C$_4$H$_{10}$ and 0.3% SF$_6$ [10]. Water vapor is added to the gas mixture in order to maintain an absolute humidity of about 8000 ppm (relative humidity ~ 37%) and avoid any changes in the resistivity of the electrodes [11]. The gas gap thickness uniformity is ensured by cylindrical polycarbonate spacers, placed on a $10 \times 10$ cm$^2$ grid. The inner surface of the bakelite, which is in contact with the gas mixture, is coated with a double linseed oil layer [12], while the outer surface is coated with a conductive graphite paint. One of the graphite layers is connected to the high voltage (HV), whereas the other one is connected to the ground, in order to apply a uniform electric field across the gas gap.
Each RPC is read out with copper strips on both sides in order to obtain two-dimensional position information. The pitch and length of the read-out strips vary within each station to keep the occupancy of the detector constant. During the LHC Run 1 (2010–2013) and Run 2 (2015–2018), the RPCs were equipped with the ADULT front-end electronics (FEE) [13], which has no amplification and a minimum discrimination threshold of ∼ 7 mV. The ADULT FEE allows one to operate the detectors in the so-called maxi-avalanche mode [14–16], with the gas mixture described above and a working HV between ∼ 10.0 and ∼ 10.4 kV [17].

The RPC detectors have shown stable operation and an efficiency greater than 98% from the beginning of Run 1 up to now [18]. During the LHC Run 3 (2021 onwards), ALICE will take data in Pb-Pb collisions at an instantaneous luminosity ∼ 6 times higher than in Run 2, thanks to a major upgrade of both the LHC injectors and the experiment [19]. In view of the harsher running conditions, a new amplified FEE, called FEERIC [20], has been developed. This will allow one to operate the RPCs in pure avalanche mode, with a lower charge per hit (by a factor 3÷5), hence improving the rate capability and reducing ageing effects. Thanks to the amplification of signals, the working HV values of ALICE-muon RPCs equipped with FEERIC are expected to be ∼ 500 V lower than the ones with ADULT FEE [21]. One RPC of the ALICE muon identification system has been equipped with FEERIC prototypes since the beginning of Run 2, with fully satisfactory performance [21].

The current ALICE mixture is not environment-friendly because of the presence of greenhouse gases. In this paper we report the results of the R&D studies on eco-friendly gas mixtures. A reasonable goal could be to operate the change of gas mixture between the LHC Run 3 and Run 4, during the LHC Long Shutdown 3 (2024–2026).

The paper is organized as follows. In section 2 we discuss the approach to find alternative, environment-friendly, gas mixtures. The experimental set-up for the R&D studies is presented in section 3, whereas the results of these tests are reported in section 4. Finally, conclusions are drawn in section 5.

2 Search for environment-friendly gas mixtures

A greenhouse gas is classified according to its Global Warming Potential (GWP), which is a relative measurement of how much heat is trapped into the atmosphere with respect to the same mass of CO₂. The GWP of each gas is determined by two key parameters. The first parameter, the radiative efficiency, indicates the capability of the gas to absorb energy in the atmosphere. For each gas, this value depends on the absorption of infrared radiation and the position in the spectrum of the absorbing wavelengths. The second important parameter is the atmospheric lifetime, which denotes how long the gas stays in the atmosphere.

Fluorinated gases may generally have very high GWPs, because their absorption of infrared radiation is high and they stay in the atmosphere for very long periods. On the contrary, hydrocarbons have generally lower GWPs than fluorinated gases as their infrared absorption is not so much efficient, even if their lifetimes are similar to the majority of fluorinated gases. In fact, the gas mixture of the ALICE muon RPCs features C₂H₂F₄ (GWP ∼ 1430) and a small concentration of SF₆ (GWP ∼ 22800), while the GWP of i-C₄H₁₀ is only 3 [22]. The current ALICE mixture has an overall GWP of 1351, almost 95% of which is due to the presence of C₂H₂F₄.
Recent regulations [22] from the Europian Union (EU) impose the reduction of the emissions of fluorinated greenhouse gases (F-gases) in EU countries since January 2015. The major EU provision is a gradual phase out of the hydrofluorocarbons (such as C$_2$H$_2$F$_4$) available on the market in order to limit the total amount of the production. The phasing out of F-gases could lead to a progressive increase of their price due to the limited future availability.

Concerning the operation of the ALICE muon identification system in the forthcoming data taking periods, the limited future availability of C$_2$H$_2$F$_4$ needs to be carefully considered as well as the possible increase of its price in the coming years. In addition, CERN has elaborated a number of strategies to reduce as much as possible the greenhouse gas emissions or, at least, optimize their usage in the LHC experiments [23]. For these reasons, R&D studies on eco-friendly gas mixture for the ALICE-muon RPCs are envisaged.

One of the possible approaches relies on the replacement of C$_2$H$_2$F$_4$ with a chemically similar molecule [24–26], but with a low enough GWP. The tetrafluoropropene C$_3$H$_2$F$_4$ is similar to C$_2$H$_2$F$_4$. Unlike C$_2$H$_2$F$_4$, the tetrafluoropropene C$_3$H$_2$F$_4$ is an olefin, so it presents a double bond in the chain of carbon atoms. The atmospheric lifetime of C$_3$H$_2$F$_4$ is estimated to be ~ 2 weeks [27], thus its GWP turns out to be extremely lower than that of C$_2$H$_2$F$_4$.

Isomers of tetrafluoropropene are HFO1234ze(Z), HFO1234ze(E) and HFO1234yf. The boiling points of these isomers at atmospheric pressure are 9.8 °C, −19 °C and −30 °C, respectively. Because of its high boiling point, HFO1234ze(Z) does not turn out to be suitable for gaseous detectors, which generally operate at room temperature and atmospheric pressure. On the contrary, HFO1234yf and HFO1234ze(E) have a lower boiling point than HFO1234ze(Z). The flammability of HFO1234yf is not excluded at room temperature while HFO1234ze(E) is not flammable below 30 °C. HFO1234ze(E) is also compatible with a large number of polymeric materials and is considered safe for refrigeration and air-conditioning applications if guidelines and safety instructions are respected [28]. The GWP of HFO1234ze(E) in the atmosphere is ~ 200 times lower than the one of C$_2$H$_2$F$_4$. Indeed, the GWP of HFO1234ze(E) is 7 according to [22], whereas latest studies have revised this value to less than 1 [29]. In addition, the environmental impact of HFO1234ze(E) by its atmospheric decay has been evaluated to be negligible [27]. For all these reasons, HFO1234ze(E) (trans-1,3,3,3-Tetrafluoroprop-1-ene) is the most interesting isomer for our purposes, even if the possibility of HFO1234ze(E) to release highly corrosive and toxic components in case of fire has to be evaluated in more detail.

We have performed R&D studies on RPCs with HFO1234ze(E)-based gas mixtures and the FEERIC electronics. The new mixture has to provide similar performance as the current one [21]. The approach to the problem is exclusively experimental, because several parameters of HFO1234ze(E), such as the electron collision cross sections, the photon absorption spectrum and the chemical reactivity, are still unknown. This prevents a more theoretical approach and the implementation of reliable simulations.

3 The experimental set-up

A dedicated experimental set-up has been used to carry out R&D studies on various gas mixtures, flushing one small-size (50 × 50 cm$^2$, 2 mm gas gap) RPC with the same features of the RPCs used in ALICE.
The detector is horizontally placed and exposed to the cosmic-ray flux. The gas mixture is prepared thanks to the use of a dedicated gas mixing unit, which can mix up to four different gases. The gas mixture composition is regulated by four mass-flow controllers (F-201CV) by Bronkhorst. One mass-flow controller, previously calibrated for C$_2$H$_2$F$_4$, has been used for HFO1234ze(E) according to the calibration curve in [30]. The mixture is humidified by bubbling it in distilled water at 10°C with the aim to obtain a gas relative humidity in the range of 30–50%. For all measurements in this work, the gas flow is set to ~ 6 volume changes per hour. The applied high voltages ($HV_{appl}$) are corrected for temperature and atmospheric pressure. All the results of this work will be given in terms of the corrected high voltage: $HV = HV_{appl} \cdot \frac{p_0}{p} \cdot \frac{T}{T_0}$, where $p$ is the pressure, $T$ is the temperature, $p_0$ and $T_0$ are reference values and are equal to 1000 mbar and 293.15 K, respectively. The cosmic-ray trigger is provided by a three-fold coincidence of plastic scintillators, coupled with photomultipliers. The trigger area is about 4 × 8 cm$^2$, while the average trigger rate is 0.12 Hz. The read-out strips have 2 cm pitch. The strip plane on the anode side of the RPC is equipped with FEERIC front-end cards, configured to discriminate negative signals.

FEERIC [20] is designed in the AMS 350 nm CMOS technology and is able to amplify signals up to ±1 pC in the linear range. Its intrinsic time resolution is lower than 1 ns (root mean square) in the input charge range 100–1000 fC, which is the typical input charge expected for the RPCs in avalanche mode.

The four adjacent strips covering the trigger area are read out on both ends: on one end the signals are discriminated by FEERIC, while on the other end the analogic signals are summed by a fan-in/fan-out module and then digitized by an oscilloscope. This allows for the simultaneous measurement of the efficiency, with FEERIC, and of the amplitude of the signals. The digital oscilloscope (LeCroy WaveSurfer 510), used for measuring the amplitude signals, has a bandwidth of 1 GHz and sampling rate of 10 GS/s.

The efficiency of the RPC detector is measured by analyzing its response in the presence of a trigger signal. If at least one strip fires, the detector is considered efficient. The threshold used for the data acquisition is ~ 130 fC (70 mV after the amplification [21]), which is the same setting used for the FEERIC prototypes installed in ALICE.

First of all, a measurement of the efficiency curve versus $HV$ has been performed with the ALICE mixture in order to characterize the detector. The results$^1$ are reported in figure 1. The maximum efficiency of the RPC with the ALICE mixture is ~ 95% in this specific experimental set-up. The working point corresponds to the beginning of the efficiency plateau. For this RPC, the plateau is reached at 9.8 kV when the detector is equipped with FEERIC and flushed with the ALICE mixture, as shown in figure 1.

During the charge multiplication in the gas gap, the high density of free charges and UV photons may induce the transition from an electron avalanche to a streamer [31], which liberates more charge in the gas and induces larger signals on the electrodes with respect to a standard

$^1$The efficiency curves as a function of $HV$ are fitted by the following sigmoid function:

$$\epsilon(HV) = \frac{\epsilon_{max}}{1 + \exp[-\alpha \cdot (HV - HV_{0.5})]}$$

(3.1)

where $\epsilon_{max}$ is the efficiency for $HV \to \infty$, $HV_{0.5}$ is the $HV$ value for which the efficiency is equal to half of $\epsilon_{max}$ and $\alpha$ is the slope at the inflection point. The voltage region $\Delta HV_{0.1\rightarrow0.9}$, where the efficiency rises from 10% to 90% of its maximum, can be easily calculated as 4.4/$\alpha$, thanks to the symmetry of the sigmoid function.
Figure 1. Efficiency curve and streamer probability of a RPC detector equipped with FEERIC front-end electronics (with threshold set to 70 mV) measured with cosmic rays. The solid line is a sigmoid-function fit to the efficiency curve (see text for details). Statistical error bars are hidden by markers.

avalanche. The presence of streamers is not desirable because they imply a reduction in the rate capability of the detector caused by the large local voltage drop, which is slowly recovered because of the high resistivity of the electrodes. In addition, streamers may enhance ageing effects [12].

Figure 2 shows the amplitude of signals induced on the strips when the RPC is operated at the working point (9.8 kV) with the ALICE mixture. The amplitude spectrum of the signals is a long-tailed distribution, as shown in figure 2, and we used an amplitude threshold of 18 mV to tag streamers. The induced charge of signals with an amplitude of ~ 18 mV corresponds to ~ 3 pC. In this work, the fraction of streamers is used, along with the efficiency, to assess the performance of the tested gas mixtures. In figure 1 the streamer probability versus $HV$ is shown for the ALICE mixture. The streamer probability, evaluated with this threshold, is about 5% at the working point with the ALICE mixture.

4 Methodology and results

The direct replacement of C$_2$H$_2$F$_4$ in the ALICE mixture with 100% HFO1234ze(E) is not advisable because it leads to a too high RPC working point. For example, an RPC operated with 100% HFO1234ze(E) is completely inefficient up to 14 kV, which is too close to the maximum applicable $HV$ value for the ALICE RPCs. Indeed, the maximum $HV$ value that the power supply used for the RPCs of the ALICE muon identification system can provide is 15 kV. Similarly, $HV$ cables and connectors are compatible with voltages up to 15 kV.

In order to lower the working point, we carried out many tests with addition of Ar, CO$_2$, O$_2$ and N$_2$ in the mixture at different concentrations. CO$_2$ turned out to be the most promising solution.

In this section the results of the tests on HFO1234ze(E)-based mixtures with the addition of CO$_2$ are shown. All gas mixtures presented in this paper are made of four components. Since the contribution of each gas in the mixtures is complex to investigate, we proceeded by changing the fractions of two gas components out of four at a time, evaluating how their ratio affects the performance of the RPC.
The dashed line illustrates the amplitude threshold of 18 mV.

4.1 Variation of the ratio between HFO1234ze(E) and SF$_6$

The role of SF$_6$ in reducing the avalanche size and suppressing streamers in C$_2$H$_2$F$_4$-based mixtures is well known in literature (see e.g. [16]). In order to study the suppression of streamers in HFO1234ze(E)-based mixtures, we firstly evaluated how the percentage of SF$_6$ affects the RPC performance, while the concentrations of CO$_2$ and i-C$_4$H$_{10}$ are kept constant. Figure 3a shows that the working point of the RPC detector increases with the addition of SF$_6$. A small variation on the concentration of SF$_6$ leads to an important effect on the working point: the shift of the working point is $\sim$ 500 V from 0.3% to 1.0% SF$_6$. In figure 3b the impact of the SF$_6$ percentage on the streamer probability is shown as a function of $HV - HV_{\varepsilon=0.9}$. $HV_{\varepsilon=0.9}$ is the voltage where the efficiency is 90% of its maximum value ($\varepsilon_{\text{max}}$), and is obtained, for each mixture, by fitting the efficiency curve with a sigmoid function.

No significative changes in the streamer suppression are observed if SF$_6$ is increased from 0.3% to 0.6%, while the streamer suppression is slightly better with 1.0% SF$_6$. In fact, the values of streamer probability with 0.3% and 0.6% SF$_6$ are higher than 10% at $HV - HV_{\varepsilon=0.9}$ of 200 V, while the streamer fraction decreases to $\sim$ 6% with 1.0% SF$_6$. The values of streamer probability with 1.0% SF$_6$ are lower than the ones with 0.3% and 0.6% SF$_6$ for all values of $HV - HV_{\varepsilon=0.9}$ greater than 0 V.

Since the streamer probability with 1.0% SF$_6$ is similar to the one of the ALICE mixture, hereafter we investigate the contribution of HFO1234ze(E), CO$_2$ and i-C$_4$H$_{10}$ while keeping always constant the fraction of SF$_6$ at 1.0%.

4.2 Variation of the ratio between HFO1234ze(E) and CO$_2$

In order to study the effects of HFO1234ze(E) and CO$_2$ on the efficiency of the RPC and the fraction of streamers, we analysed three different cases: without i-C$_4$H$_{10}$, with 10% i-C$_4$H$_{10}$ and
20% $i$-$C_4H_{10}$, while the fraction of SF$_6$ is always 1%. If the proportion of CO$_2$ is increased and that of HFO1234ze(E) is decreased, the working point turns out to be shifted towards lower voltages, as shown in figure 4, where the fraction of $i$-$C_4H_{10}$ is 10%. In the same figure, the streamer probability as a function of $HV$ is shown. The streamer probability for the gas mixture 55.5% CO$_2$, 33.5% HFO1234ze(E), 10% $i$-$C_4H_{10}$, 1.0% SF$_6$ is $\sim$ 40% at the working point, whereas values of streamer probability are $\sim$ 20% at the working point for the other two gas mixtures in figure 4.

A similar trend of the working point position as a function of the HFO1234ze(E) fraction has been observed without $i$-$C_4H_{10}$ and with 20% $i$-$C_4H_{10}$, as reported in figures 5a and 5b. In the same figures, the streamer probability as a function of $HV$ is also shown.

### 4.3 Variation of the ratio between HFO1234ze(E) and $i$-$C_4H_{10}$

In order to explore the effects of $i$-$C_4H_{10}$ in the mixture, we evaluated the RPC performance at different ratios between HFO1234ze(E) and $i$-$C_4H_{10}$ (figure 6). In particular, we compared the performance of the detector with the gas mixture 50% CO$_2$, 39% HFO1234ze(E), 10% $i$-$C_4H_{10}$, 1% SF$_6$ and with the gas mixture 50% CO$_2$, 29% HFO1234ze(E), 20% $i$-$C_4H_{10}$, 1% SF$_6$, already presented in figures 4 and 5b, respectively. The working point of the detector is shifted towards
Figure 4. Efficiency and streamer probability for gas mixtures with different ratios between C$_3$H$_2$F$_4$ (HFO1234ze(E)) and CO$_2$, while the concentrations of $i$-C$_4$H$_{10}$ and SF$_6$ are kept constant (10% and 1%, respectively).

Figure 5. Efficiency and streamer probability for gas mixtures without $i$-C$_4$H$_{10}$ (a) and with 20% $i$-C$_4$H$_{10}$ (b) for different ratios between C$_3$H$_2$F$_4$ (HFO1234ze(E)) and CO$_2$, while the concentration of SF$_6$ is kept constant at 1%.
lower voltages if the \( i-C_4H_{10} \) is increased and HFO1234ze(E) is decreased. In the same figure, the streamer probability as a function of \( HV \) is shown. Streamer fractions are \( \sim 20\% \) at the working point for both gas mixtures.

The increase of the working point with the increase of the ratios HFO1234ze(E)/CO\(_2\) (figures 4 and 5) and HFO1234ze(E)/\( i-C_4H_{10} \) (figure 6) might be explained by the electron attachments to HFO1234ze(E) [32]. Indeed, the effect of the electron attachment turns out to be dependent on the partial pressure of the HFO1234ze(E) at the atmospheric pressure, as shown by the measurements of its electron swarm parameters [32]. Therefore the concentration of this gas strongly affects the total gain of the gas mixture.

4.4 Variation of the ratio between CO\(_2\) and \( i-C_4H_{10} \)

Effects of the concentration of \( i-C_4H_{10} \) have been carefully studied because of its impact on the gas mixture flammability. The reduction, or even better the removal, of this flammable component in the gas mixture would be desirable for safety and practical reasons. On the other hand, the increase of \( i-C_4H_{10} \) against HFO1234ze(E) implies a significant shift of the working point towards lower \( HV \) values. In fact, the increase of the \( i-C_4H_{10} \) fraction from 10% to 20% reduces the working point by \( \sim 600 \) V, as shown in figure 6. In this section, the substitution of \( i-C_4H_{10} \) with CO\(_2\) has been investigated.

Figure 7a shows the efficiency curve at different ratios between \( i-C_4H_{10} \) and CO\(_2\), while the concentrations of HFO1234ze(E) and SF\(_6\) are kept constant (about 34% and 1% respectively). The fraction of streamers is shown in figure 7b as a function of \( HV - HV_{e=0.9} \).

The shift of the working points is not monotonic versus the ratio CO\(_2\)/\( i-C_4H_{10} \). The working point clearly increases (by \( \sim 300 \) V) when going from 10% to 20% \( i-C_4H_{10} \), while no clear trend emerges when the \( i-C_4H_{10} \) fraction is varied between 0% and 10%. In this range, the variations of the working point are anyway small. The fraction of streamers does not appear to be significantly reduced with the increase of \( i-C_4H_{10} \) in place of CO\(_2\), as shown in figure 7, except for the fact that the streamer probability rises slightly less steeply with 15% \( i-C_4H_{10} \) than with lower fractions of...
Figure 7. Efficiency (a) and streamer probability (b) for gas mixtures with different ratios between $i$-$C_4H_{10}$ and CO$_2$, while the concentrations of C$_3H_2F_4$ (HFO1234ze(E)) and SF$_6$ are kept almost constant.

Table 1. Values of voltage region $\Delta HV_{0.1 \rightarrow 0.9}$, where the efficiency rises from 10% to 90%, at different ratios between CO$_2$ and $i$-$C_4H_{10}$.

| Gas mixture                                   | $\Delta HV_{0.1 \rightarrow 0.9}$ [kV] |
|-----------------------------------------------|---------------------------------------|
| 65.5% CO$_2$, 33.5% HFO1234ze(E), 0.0% $i$-$C_4H_{10}$, 1.0% SF$_6$ | 1.14 ± 0.06                           |
| 60.5% CO$_2$, 33.5% HFO1234ze(E), 5.0% $i$-$C_4H_{10}$, 1.0% SF$_6$ | 1.23 ± 0.07                           |
| 55.5% CO$_2$, 33.5% HFO1234ze(E), 10.0% $i$-$C_4H_{10}$, 1.0% SF$_6$ | 1.07 ± 0.05                           |
| 50.0% CO$_2$, 34.0% HFO1234ze(E), 15.0% $i$-$C_4H_{10}$, 1.0% SF$_6$ | 0.70 ± 0.03                           |
| 44.5% CO$_2$, 34.5% HFO1234ze(E), 20.0% $i$-$C_4H_{10}$, 1.0% SF$_6$ | 0.84 ± 0.04                           |

this gas. Finally, we observe that the voltage region ($\Delta HV_{0.1 \rightarrow 0.9}$), where the efficiency rises from 10% to 90%, becomes smaller as the $i$-$C_4H_{10}$ fraction increases and the CO$_2$ fraction decreases, as reported in table 1.

The interplay of $i$-$C_4H_{10}$ and CO$_2$ in HFO1234ze(E)-based mixtures turns out to be rather complex and needs further studies in order to explore the feasibility of RPC mixtures without flammable components.
4.5 Promising gas mixtures with low GWP

Eco-friendly gas mixtures with similar features as the current ALICE gas mixture have been selected as promising candidates to replace the current mixture in the future. The parameters used to identify a promising gas mixture are the position of the working point and the streamer probability. The working point should not be too much larger than the working point of the ALICE-muon RPCs. The fraction of streamers should be possibly not larger than the one of the current ALICE gas mixture. However, higher streamer fractions may be acceptable provided it can be shown that they do not compromise the performance and stability of the ALICE muon identification system. Of course, the GWP of the promising gas mixtures must be lower than that of the ALICE RPC gas mixture.

Figure 8 shows the most promising gas mixtures, among those tested so far:

- 50% CO$_2$, 39.7% HFO1234ze(E), 10% $i$-C$_4$H$_{10}$, 0.3% SF$_6$ and 50% CO$_2$, 39% HFO1234ze(E), 10% $i$-C$_4$H$_{10}$, 1% SF$_6$. Their GWPs are respectively 72 and 232, while the GWP of the current ALICE mixture is 1351. For both mixtures, the dark current at the working point is stable in time and not significantly different from that measured with the standard mixture. The cluster size and the signal jitter with respect to the scintillator trigger are also very similar for the three mixtures.

As shown in figure 8a, the working point of the detector with the first mixture (50% CO$_2$, 39.7% HFO1234ze(E), 10% $i$-C$_4$H$_{10}$, 0.3% SF$_6$) is quite close to the working point of the ALICE RPCs during LHC Run 1 and Run 2, which should be safe for operation, while the streamer probability is not as low as in the current ALICE mixture, due to the smaller fraction of SF$_6$. On the contrary, the working point with the second mixture (50% CO$_2$, 39% HFO1234ze(E), 10% $i$-C$_4$H$_{10}$, 1% SF$_6$) is $\sim$ 1.5 kV higher than the one of the current ALICE mixture, but the suppression of streamers is higher than the first promising gas mixture. In fact, the streamer probability with 0.3% SF$_6$ is $\sim$ 12% at $HV - HV_{e=0.9}$ of $\sim$ 200 V, while it decreases to $\sim$ 6% with 1.0% SF$_6$, as reported in figure 8b.

5 Conclusions and outlook

Studies on HFO1234ze(E)-based mixtures have shown a strong dependence between the working point of the detector and the concentration of HFO1234ze(E), most probably due to its electron attachment. Promising results are obtained with the addition of CO$_2$ in a HFO1234ze(E)-based mixture.

SF$_6$ turns out to play a crucial role in suppressing streamers in HFO1234ze(E)-based mixtures, as was the case for C$_2$H$_2$F$_4$-based mixtures.

The increase of $i$-C$_4$H$_{10}$ in place of CO$_2$ does not seem to play a crucial role in reducing the streamer probability, which is promising in light of a possible future reduction or removal of this (flammable) gas from the ALICE mixture. On the contrary, the reduction of $i$-C$_4$H$_{10}$ against CO$_2$ in the mixtures studies so far leads to a less sharp efficiency turn-on, which calls for more effort in this direction.

The most promising gas mixtures, which have been found so far, consists of 50% CO$_2$, 39.7% HFO1234ze(E), 10% $i$-C$_4$H$_{10}$, 0.3% SF$_6$ and 50% CO$_2$, 39% HFO1234ze(E), 10% $i$-C$_4$H$_{10}$, 1% SF$_6$. The GWPs of these two gas mixtures are significantly lower than the GWP of the current ALICE mixture.
Figure 8. Efficiency (a) and streamer probability (b) for the standard ALICE mixture and the most promising C₃H₂F₄-based gas mixtures.

Measurements of the RPC performance under irradiation, e.g. at the CERN Gamma Irradiation Facility [33], will be crucial for assessing the rate capability and ageing properties resulting from the considered mixtures, thus qualifying these for high-luminosity operation at the LHC of the ALICE muon RPCs.

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