Characterization of new eco friendly gas mixtures based on HFO for RPCs

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ABSTRACT: The Resistive Plate Chambers standard mixture, mainly based on C\textsubscript{2}H\textsubscript{2}F\textsubscript{4}, has a high Global Warming Potential (GWP) and therefore the search for RPC eco friendly gases is mandatory. In fact if some additional restrictions on greenhouse gas emissions will be imposed or if this gas becomes too expensive due to a reduction on the industrial demand, it will be impossible to use it anymore. The main problem is that the detector already installed in the experiments can not work properly with the eco-friendly gases already studied and for this reason the search for a suitable eco-friendly gas mixture is crucial. In this work we present the results on the detector performance in terms of efficiency, prompt and ionic charge, with different gas mixtures.

KEYWORDS: Gaseous detectors; Resistive-plate chambers
1 Introduction

The Resistive Plate Chambers [1] standard mixture, composed by C$_2$H$_2$F$_4$ (as the main component), i-C$_4$H$_{10}$ and SF$_6$, is presently used in several experiments with a high radiation environment. Unfortunately it has a high Global Warming Potential of about 1465 (for C$_2$H$_2$F$_4$/i-C$_4$H$_{10}$/SF$_6=94.5\%/5\%/0.5\%$), mainly due to the C$_2$H$_2$F$_4$ (SF$_6$ gives a minor contribution of $\sim114$). The use of this mixture could be problematic in the near future if additional restrictions on the greenhouse gas emissions will be imposed [2, 3] or if this gas becomes too expensive due to a reduction on the industrial demand.

The main problem in searching for alternative mixtures is to maintain good avalanche saturation properties and the separation between avalanche operation mode and streamer contamination over a large electric field range (for the standard mixture is $\sim 1\text{kV}$).

For what concerns the new generation of RPCs that are planned for the ATLAS Phase-II upgrade [4], the new detector layout together with the development of new Front-End electronics give the possibility to work with alternative mixtures with lower GWP [5–9], based on HFO, CO$_2$, i-C$_4$H$_{10}$ and SF$_6$. Instead the RPCs already installed in the experiment can not be upgraded and therefore they can not work with these mixtures. With HFO-based new gas mixtures, at full efficiency working point the avalanches are wider, carry more charge and the transition to streamers passes through multi-avalanches events with a high charge content. The charge distributions are broader and the pure saturated-avalanche regime can be identified only when the efficiency is still too low.

The goal of this study is to identify the best new eco-friendly gas mixture for generic RPCs and to optimize the concentrations of the various components inside the mixture.
2 Experimental setup

A schematic view of the RPC configuration is shown in figure 1.

![Schematic view of the RPCs configuration.](image)

Figure 1. Schematic view of the RPCs configuration.

An RPC with 2 mm gas gap and two 1.8 mm thick electrodes (ATLAS-like) has been used in this test. The detector dimensions are $57 \times 10 \text{ cm}^2$. The prompt induced signal is read out on both sides of a single strip line with the oscilloscope without signal amplification. One of the two signals is acquired with the maximum sensibility of the oscilloscope to optimize the analysis for the avalanche, while the other is acquired with a variable scale to study the streamers. The oscilloscope has a bandwidth of 3 GHz and a sampling velocity of $10 \ G\text{s/s}$ and the time window acquired for the prompt signal is 200 ns.

The ionic signal is read out on a resistance of about $10 \text{k}\Omega$ on the ground graphite electrode over a time window of 100 $\mu$s. This signal represents the best estimate of the total charge associated with the single event.

The trigger system consists of four RPCs (see figure 1) and a 1 mm gas gap RPC has been used as time reference and confirmation for the efficiency measurement.

The mixtures have been prepared using four mass flow meters properly calibrated. The mixture humidity corresponds to the ambient humidity, which enters into the mixture in the last part of the plastic tube.

3 Analysis parameters definition

A signal is considered efficient if it crosses an amplitude threshold, defined as the 5*Root Mean Square (RMS) of the background amplitude distribution, which is measured in a time window of 20 ns just before the trigger signal. No Front-End electronics has been used, and the lowest equivalent threshold is $2.2 \text{mV}$ (highest oscilloscope sensitivity), which is substantially higher than the one of the ATLAS RPCs ($\sim 1.2 \text{mV}$).

The avalanche charge, the total prompt charge and the ionic charge are measured for each acquired waveform. The avalanche charge is defined as the integrated charge in 10 ns around the first peak. The total prompt charge is the integrated charge in the whole time window of 180 ns (the first 20 ns are used for the background measurement), so it includes also the multi-avalanches and the streamers contributions. The ionic charge is measured in the full time window of 80 $\mu$s after trigger. The window of 20 $\mu$s before the trigger has been used for the background measurement.

The delivered charge in HFO-based mixtures takes an important contribution from multi-avalanches (see figure 2), therefore, instead of streamer, we define an extra-charge, which includes also streamers, due to all signals observed in 180 ns.
A threshold of 20 pC has been fixed to discriminate what we call an *extra-charge* event, which represents a charge value that is 10 times that of the mean avalanche charge.

![Avalanche](avalanche.png) ![Multi avalanche](multi_avalanche.png)

**Figure 2.** Example of an avalanche event (left) and a multi avalanche event (right).

4 Measurements strategy and experimental results

In order to study the properties of the different gas components, a series of measurements have been performed starting from the mixture composed by C₃H₂F₄ (HFO-1234ze)/CO₂/i-C₄H₁₀/SF₆ =38/56/5/1. In the first series the CO₂/SF₆ ratio has been kept constant, while HFO/i-C₄H₁₀ concentrations have been changed. In the second series the i-C₄H₁₀/SF₆ fraction is fixed and the concentrations of the main components of the mixture vary.

Finally, a different new type of HFO, C₃H₂ClF₃ (HFO-1233zd) [10], has been tested.

The performance of each gas mixture has been studied in terms of efficiency, avalanche-extra charge separation and total charge delivered in the detector. For all measurements the standard mixture (C₂H₂F₄/i-C₄H₁₀/SF₆/=94.5/5/0.5) has been kept as a reference. In figure 3 the results for the standard mixture are summarized.

The efficiency curve shows that the voltage gap between the efficiency plateau and the streamer appearance is of ~ 1 kV (figure 3 (a)). Figure 3 (b) shows the ionic charge, total prompt charge and avalanche charge as a function of the high voltage.

4.1 Mixtures with HFO1234ze/CO₂ /i-C₄H₁₀/SF₆

The first measurement has been performed with the mixture composed by HFO1234ze/CO₂/i-C₄H₁₀/SF₆ = 38/56/5/1, with the CO₂ as the main component.

In general we would like to work with as much CO₂ as possible, because the HFO plays the role of *quencher* [5] and it moves the working point towards very high voltages. A greater fraction of HFO is expected to show lower extra charge probability and lower current flowing inside the detector.

Two sets of measurements have been performed, one to study the quenching components and the other to study the ratios between the main components of the mixture.
4.1.1 Performance with HFO1234ze/ i-C\textsubscript{4}H\textsubscript{10} variable ratio

In this section the CO\textsubscript{2} and SF\textsubscript{6} concentrations are fixed at the ratio 56/1, while the HFO concentration is lowered in favour of i-C\textsubscript{4}H\textsubscript{10}. Three mixtures are studied: HFO/i-C\textsubscript{4}H\textsubscript{10} = 38/5, 33/10, 29/14.

The results are reported in figure 4.

We can notice that the addition of 9% of i-C\textsubscript{4}H\textsubscript{10} lowers the operating voltage of about 600 V if compared with the mixture with 5% of i-C\textsubscript{4}H\textsubscript{10} and the avalanche-extra charge separation is ~ 600 V between the three mixtures (figure 4.a-4.b). If we look at the ionic charge, which represents the total charge delivered inside the detector, we can see that the average value is the same (~ 30 pC) for the three mixtures at the respective working points (figure 4.c): 11.6 kV (14% of i-C\textsubscript{4}H\textsubscript{10}), 11.9 kV (10% of i-C\textsubscript{4}H\textsubscript{10}) and 12.1 kV (5% of i-C\textsubscript{4}H\textsubscript{10}). The same happens for the total prompt charge and takes the value of ~ 4 pC at the respective working points (figure 4.d).

In conclusion the decrease of operating voltage is only due to the decrease of HFO concentration and there is no effect in increasing i-C\textsubscript{4}H\textsubscript{10} in terms of avalanche-extra charge separation and total charge delivered at the working point.

4.1.2 Performance with HFO1234ze/CO\textsubscript{2} variable ratio

In this section the fraction i-C\textsubscript{4}H\textsubscript{10}/SF\textsubscript{6} has been kept constant to the ratio 5/1 and the concentration of CO\textsubscript{2} has been increased in steps of 5%. The reported HFO/CO\textsubscript{2} ratios are 38/56, 33/61 and 28/66.

In figure 5 the results are reported. It is visible that the reduction of HFO moves the working point to lower voltages of about 400 V every 5% of HFO reduction (figure 5.a). For what concerns the avalanche-extra charge separation, the range is larger in the mixture with more HFO (figure 5.b). The total prompt charges for the three gas mixtures at the working point (11.3 kV for the ratio 28/66, 11.7 kV for the ratio 33/61 and 12 kV for the ratio 38/56) are consistent with each other (~ 3 pC), while the total ionic charge is higher in the mixture with more HFO (~ 30 pC against ~ 20 pC for the other two) as you can see in figures 5.c-5.d.
Figure 4. Efficiency (a), extra charge probability (b), ionic charge (c) and total prompt charge (d) as a function of the high voltage at fixed 56/1 CO$_2$/SF$_6$ ratio for the three mixtures: HFO/i-C$_{4}H_{10}$ = 29/14 (red points), 33/10 (blue squares) and 38/5 (black triangles).

4.2 Mixtures with HFO1233zd/CO$_2$/i-C$_{4}H_{10}$/SF$_6$

In this section the results related to a different type of HFO, the HFO1233zd [10], are reported. This molecule, C$_3$H$_2$ClF$_3$, has an extra Chlorine and one Fluorine less with respect to the HFO-1234ze. Similar molecules, containing two different halogen atoms, were already tested as RPC gas components. CF$_3$Br was shown to have a relevant effect in reducing the streamer size in RPC operating in streamer mode [15]. Moreover, the pure avalanche mode without any streamer contamination was first observed in RPCs filled with pure CF$_3$Br. More recently, the CF$_3$I molecule was successfully tested as an alternative RPC gas [8]. Unfortunately, none of these molecules is suitable for very large size RPC systems, which require not-flammable and not toxic gases. Indeed CF$_3$Br has a very high ozone depletion potential and CF$_3$I is very poisonous.

C$_3$H$_2$ClF$_3$ is an industrial gas and does not have the above problems and the test made with the other two-halogen molecules (CF$_3$Br and CF$_3$I) suggests that it could be suitable to suppress the streamer appearance and to keep the avalanche size to modest values. We tried to start with the starting point mixture (HFO/CO$_2$/i-Butane/ SF$_6$=38/56/5/1) and replace the HFO1234ze with HFO1233zd. With this HFO1233zd concentration the first efficient signal has been observed at 14 kV. The concentration of HFO1233zd has been therefore drastically reduced.
Figure 5. Efficiency (a), extra charge probability (b), ionic charge (c) and total prompt charge (d) as a function of the high voltage at fixed $5/1$ $i$-$C_4H_{10}/SF_6$ ratio for the three mixtures: HFO/$CO_2 = 28/66$ (blue squares), 33/61 (red points) and 38/56 (black triangles).

A test with a new gas mixture composed by HFO-1233zd/$CO_2$ /$i$-$C_4H_{10}$ with and without the presence of $SF_6$ has been performed, in order to study if this gas could give the same benefits of the $SF_6$.

### 4.2.1 Performance with mixtures with and without $SF_6$

The mixtures under study are composed by HFO-1233zd/$CO_2$ /$i$-$C_4H_{10}$=15/80/5 with and without the addition of 1% of $SF_6$. The experimental results are reported in figure 6.

It can be seen that the mixture without $SF_6$ has an operating voltage $\sim$ 300–400 V (at 90% efficiency) lower and shows a steeper rise respect to the mixture with $SF_6$. This is an expected behaviour [12] and can be explained with the very high electronegativity of this molecule. In fact the simultaneous presence of the HFO and $SF_6$ breaks the avalanche growth and makes harder the rise of the efficiency curve.

The total prompt charge at the plateau knee (11800 V for the mixture without $SF_6$ and 12100 V for the mixture with $SF_6$) is higher in the mixture with the $SF_6$ ($\sim$ 6 pC against 8 pC) and the same is true for the ionic charge delivered in the detector ($\sim$ 40 pC against 60 pC). Therefore, when $SF_6$ is added, it finds a very electronegative environment and the resulting effect is the premature break of the avalanche growth and not the streamers suppression.
Figure 6. Efficiency (a), extra charge probability (b), ionic charge (c) and total prompt charge (d) as a function of the high voltage for the two mixtures: HFO1233zd/CO$_2$/i-C$_4$H$_{10}$ without SF$_6$ (red points) and with 1% of SF$_6$ (blue squares).

Figure 7. Total prompt charge distribution at 24% (a), 81% (b) and 94% (c) of efficiency for the standard mixture (blue) and HFO1233zd/CO$_2$/i-Butane Mixture (red).

Figure 7 shows the comparison between the total prompt charge distributions of the standard mixture and the HFO1233zd/CO$_2$/i-C$_4$H$_{10}$ mixture at different values of the efficiency.

At lower efficiency the two distributions are concentrated near the ideal line which separate the signal charge and the background charge. When efficiency rises the total prompt charge distribution for the standard mixture shows the typical trend of saturated avalanche regime, on the opposite the HFO1233zd mixture has a broader distribution, causing an higher average working prompt charge value (see figure 7(c)).
5 Conclusions

In this work we performed a systematic performance study of a small size RPC operated with four- components gas mixture $\text{C}_3\text{H}_2\text{F}_4(\text{HFO}1234\text{ze})/\text{CO}_2$/i-$\text{C}_4\text{H}_{10}/\text{SF}_6$. The following different proportions have been studied: 29/56/14/1, 33/56/10/1, 38/56/5/1, 28/66/5/1 and 33/61/5/1. We obtained the following results:

- Decreasing HFO in the HFO/i-Butane ratio produces a decrease of the operating voltage but does not change the delivered charge, which is the same in the three mixtures and has a value of $\sim 30\, \text{pC}$. Also the avalanche- extra charge separation is the same in the three mixtures;

- Decreasing the HFO in the HFO/ CO$_2$ ratio has the effect to decrease both the operating voltage and the delivered charge. The voltage decrease is $\sim 400\, \text{V}$ every 5% of HFO reduction and the delivered charge decrease is $\sim 10\, \text{pC}$ comparing the mixture with more HFO with the mixture with less HFO. This suggests that HFO concentration could be lowered below the 28% value tested in this study. Less HFO means less Flourine molecules in the mixtures and a slower detector ageing. The avalanche-extra-charge separation range is larger in the mixture with more HFO;

We tested also another HFO gas, $\text{C}_3\text{H}_2\text{ClF}_3$ (HFO1233zd), that has never been tested before. This new component has much stronger quenching power than the previous HFO and this suggests that it might replace the SF$_6$. However this possibility requires more studies.

In general, in HFO-based gas mixtures, the saturated avalanche regime we observed in the standard one is achieved only when the efficiency is still too low. The charge distributions at high efficiency are broad because of the premature extra-charge signal appearance. The total charge delivered inside the detector and the total prompt charge in the HFO-based mixture are higher than those of the standard mixture.

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