ABSTRACT: Due to their simplicity and comparatively low cost, Resistive Plate Chambers are gaseous detectors widely used in high-energy and cosmic rays physics, when large detection areas are needed. However, the best gaseous mixtures are currently based on tetrafluoroethane, which has the undesirable characteristic of a large Global Warming Potential (GWP) of about 1400 and, because of this, it is currently being phased out from industrial use. Tetrafluoropropene (which has a GWP close to 1) is being considered as a possible replacement. Since tetrafluoropropene is more electronegative than tetrafluoroethane, it has to be diluted with gases with a lower attachment coefficient in order to maintain the operating voltage close to 10 kV. One of the main candidates for this role is carbon dioxide. In order to ascertain the feasibility and the performance of...
tetrafluoropropene-CO2 based mixtures, an R&D program is being carried out within the ALICE collaboration, employing an array of 72 Bakelite RPCs (Muon IDentifier, MID) in order to identify muons. Different proportions of tetrafluoropropene and CO2, with the addition of small quantities of isobutane and sulphur hexafluoride, have been tested with 50x50 cm$^2$ RPC prototypes with 2 mm wide gas gap and 2 mm thick Bakelite electrodes. In this contribution, results from tests with cosmic rays will be presented, together with data concerning the current drawn by a RPC exposed to the gamma-ray flux of the Gamma Irradiation Facility (GIF) at CERN.

Keywords: Resistive Plate Chambers; Eco-friendly gas mixtures; Ageing.

1. The ALICE Muon Identification System (MID)

The ALICE (A Large Ion Collider Experiment) at CERN is one of the main experiments at the Large Hadron Collider (LHC) and it is designed to study proton-proton and heavy-ion collisions (such as Pb-Pb) at ultra-relativistic energies. The main goal of ALICE is to assess the properties of the Quark Gluon Plasma (QGP), which is a state of matter where quarks and gluons are de-confined. Among the main observables used to study the QGP there is the production of heavy quarkonia, i.e. $c\bar{c}$ and $b\bar{b}$ bound states. The presence of QGP modifies the quarkonium production rates in heavy-ion collisions via the competing processes of suppression by colour screening and regeneration by quark recombination.

For this reason ALICE is equipped with a forward Muon Spectrometer which detects quarkonia via their di-muon decay channel in the rapidity interval $2.5 < \eta < 4$. It is composed of a hadron absorber, a dipole magnet, a five-station tracking system and a Muon IDentifier (MID) placed downstream a 120 cm thick iron wall.

The MID system is composed of 72 Resistive Plate Chambers (RPCs), which are position-sensitive gaseous detectors with a spatial resolution of the order of few mm and time resolution of the order of ns. The MID RPCs are arranged in two stations of two planes each. The two stations are situated at 16.1 m and 17.1 m from the Interaction Point (IP). Each station consists of 18 RPC modules and covers an area of $5.5 \times 6.5$ m$^2$ with a 1.2 x 1.2 m$^2$ central hole to accommodate the beam pipe and its shielding. A single RPC size is $70 \cdot 270$ cm$^2$ and there are three different geometry shapes: Long ($L$), Cut ($C$) and Short ($S$). Each RPC is equipped with orthogonal copper X-Y read-out strips in order to collect spatial information along both $x$ and $y$ directions, on the plane perpendicular to the beam axis. There are $\sim 21$ k strips, with pitches (1, 2 and 4 cm wide) and lengths increasing with their distance from the beam axis, in order to provide an almost flat occupancy throughout all the plane surface and to keep the momentum resolution roughly constant (see Fig. 1).

1.1. ALICE MID Resistive Plate Chambers

The ALICE MID RPCs are single-gap (2 mm thick) detectors with resistive bakelite electrodes (2 mm thick, $\rho \simeq 3 \cdot 10^9 - 1 \cdot 10^{10}$ $\Omega$ cm). The detector installation in the ALICE cavern took place in 2007 and the RPCs have been operating since
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Fig. 1. A schematic view of the ALICE MID (left) and detector composition of an half plane (right). L, C and S refer to the different geometries of the RPCs: L1, L2 and L3 indicate the Long RPCs, C the Cut ones and S the Short one. The colours indicate the different strip segmentation.

the beginning of the Run 1 in 2010. During Run 1 and Run 2, the signal was discriminated by a front-end electronics with a threshold value of 7 mV and without any pre-amplification stage.

The ALICE RPCs worked with an effective applied HV of about 10.2 - 10.5 kV at 970 mbar of pressure and 20°C. They worked in the so-called maxi-avalanche mode with an average charge per hit of 100 pC, while the maximum rate capability was about 100 Hz/cm².

From Run 3 onwards, to cope with the increased collision rate, the ALICE MID RPCs will work in avalanche mode in order to reduce the charge and increase the rate capability. This will be possible thanks to new front-end electronics (FEERIC ASIC) which includes a pre-amplification stage of the analogue signal before discrimination, with a $Q_{\text{induced}}$ threshold of 130 fC. The effective HV applied will be between 9.7 kV and 10 kV.

Regarding the gas mixture, during Run 3 the RPCs will be flushed with the same gas mixture as that used during Run 1 and Run 2: 89.7% tetrafluoroethane ($\text{C}_2\text{H}_2\text{F}_4$, R134a) 10% isobutane ($\text{i-C}_4\text{H}_{10}$), and 0.3% sulphur hexafluoride ($\text{SF}_6$). The mixture is humidified at 35-40% RH in order to avoid any variations in the resistivity of the bakelite electrodes. This gas mixture is flammable because of the presence of isobutane, while tetrafluoroethane and sulphur hexafluoride are greenhouse gases, making the current gas mixture used for the RPCs not environmentally friendly. Although the total gas volume of the MID is small (~ 0.3 m³), a more environmentally friendly gas mixture would be welcomed, so R&D studies on mixtures with lower GWP have been performed (and are still ongoing) and the results obtained up to now are shown in this paper.
2. Search for environment-friendly gas mixture

2.1. Reasons for R&D studies

European regulations^21 have imposed a reduction of the emission into the atmosphere of fluorinated Greenhouse Gases (GHG), e.g. tetrafluoroethane. This leads to a driving up of the procurement costs. Therefore, CERN is pushing for a search for more environmentally friendly gas mixtures for particle detectors, in order to limit the GHG emissions in atmosphere, and also for economic reasons. Tetrafluoroethane and sulphur hexafluoride are greenhouse gases and they are classified according to their Global Warming Potential (GWP), which is the measure of how much energy the emissions of 1 ton of a gas will absorb over a given period of time, relative to the emissions of 1 ton of carbon dioxide (CO₂). The GWP₁₀₀yr of tetrafluoroethane is 1300, while that of SF₆ is 23500^10. Considering that the GWP₁₀₀yr of isobutane is 3, and taking into account the percentage of each gas in the mixture, the GWP of the current ALICE gas mixture is equal to 1351.

The ALICE MID group started to study new environmentally friendly and non-flammable gas mixtures in 2017, with a cosmic rays set-up (see subsection 2.3). In 2018 a collaboration, called ECOGAS, between the RPC groups of ALICE, CMS and ATLAS, together with the CERN gas team, was started. In 2019 the SHiP collaboration joined the team and R&D studies are still ongoing. The goal of these R&D studies is to determine a new low GWP gas mixture that provides similar detector performances in terms of efficiency and streamer probability with respect to the current RPCs gas mixture.

2.2. From tetrafluoroethane to tetrafluoropropene

Up to now work is being carried out on a replacement for tetrafluoroethane, since R134a is responsible for 95% of the total GWP of the RPC gas mixtures at the LHC.

An appropriate candidate may be found among Hydro-Fluoro-Olefin (HFO) gases,
since they have similar properties to the R134a but with a low GWP. The tetrafluoro-
propene \( \text{C}_3\text{H}_2\text{F}_4 \) (trade-name HFO-1234ze) was chosen. This gas is not
flammable at room temperature and it has a GWP lower than 1.
However, a direct replacement of tetrafluoroethane with tetrafluoropropene is not
possible, since electron capture is more dominant for \( \text{C}_3\text{H}_2\text{F}_4 \) than for \( \text{C}_2\text{H}_2\text{F}_4 \).
This would lead to an operating voltages for RPCs higher than 15 kV, which is not
advisable for our system.
Several studies have been carried out with the addition of different gases to the
tetrafluoropropene-based gas mixture, in order to lower the HV Working Point
(WP). The most promising so far is the gas mixture with the addition of \( \text{CO}_2 \).
In this paper results will be shown on HFO-based gas mixtures with different per-
centages of \( \text{CO}_2 \), \( \text{i-C}_4\text{H}_{10} \) and \( \text{SF}_6 \).

### 2.3. Cosmic rays experimental set-up

The tests on eco-friendly gas mixtures are performed in the INFN Torino labo-
atory, using a cosmic ray test station (see figure 3).
These studies are performed with a small size ALICE RPC (50 \( \times \) 50 cm\(^2\) size, 2mm
gas gap, 16 readout strips per side with 2 cm pitch) placed horizontally and exposed
to cosmic rays. For the trigger three plastic scintillators with a total area of about
6 \( \times \) 6 cm\(^2\) are used.
The detector is equipped on one strip end with the ALICE FEERIC front-end
electronics, which amplifies and discriminates signals (threshold for the data acqui-
sition is \( \text{Q}_{\text{induced}} = 130 \text{ fC} \), 70 mV after amplification, the same as the one used for
FEERIC installed in ALICE\(^{15}\)). On the other strip end, the analogue signals are
summed with a fan-in/fan-out module and digitized by an oscilloscope. In this way
it is possible to measure the signal amplitude and charge.
The HV is applied with temperature and pressure correction\(^{a}\). Moreover, there is
the possibility to mix up to 4 different gases, using a dedicated gas mixing unit, and
the humidity of the gas mixture is kept constant at 35-40% RH in order to avoid
variations of the electrode resistivity.

### 3. Tetrafluoropropene - based gas mixture

The ALICE standard gas mixture is used as a reference, so, first of all, the efficiency
curve versus HV and the streamer probability have been evaluated by flushing the
RPC with the standard ALICE mixture (89.7% \( \text{C}_2\text{H}_2\text{F}_4 \), 10 % \( \text{i-C}_4\text{H}_{10} \) and 0.3%

\(^{a}\)All the results in this paper are given with the corrected high voltage HV:

\[
HV = HV_{\text{app}} \frac{p_0}{p} \frac{T}{T_0}
\]

where \( p_0 \) and \( T_0 \) are reference values equal to 1000 mbar and 293.15\(^{\circ}\)K respectively, while \( p \) and
\( T \) are the pressure and the temperature in the laboratory.
The plot in figure 4 shows the efficiency and the streamer probability for this mixture. As said before, the new eco-friendly gas mixture should provide similar detector performance with respect to the standard one, without impacting negatively on the detector working parameters, especially on the HV working point WP, which corresponds to the beginning of the efficiency plateau and in this case is 9.8kV. The streamer probability at the working point is 5%. The occurrence of streamers in the ALICE RPC has to be kept to a minimum because it leads both to a reduction in the rate capability and an increase in the cluster size, and may enhance ageing effects.  

The gas mixtures tested in this paper are all made up from four different gases and, since the contribution of each gas in the composition is quite complex to investigate, we change the proportions of only two gases at a time.

![Fig. 3](image1.png)

**Fig. 3.** Schematic view of the experimental set-up for R&D studies on eco-friendly gas mixtures at the INFN Torino laboratory.

![Fig. 4](image2.png)

**Fig. 4.** Efficiency curve (black dots) and streamer probability (white dots) of a RPC detector flushed with standard ALICE gas mixture. The solid line is a sigmoid-function fit to the efficiency curve.
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Fig. 5. Efficiency curves and streamer probability for gas mixtures with different ratios between C₃H₂F₄ and CO₂ (s-C₄H₁₀ and SF₆ are kept constant at 10% and 1% respectively): (i) 33.5% C₃H₂F₄ and 55.5% CO₂ (black dots), (ii) 39% C₃H₂F₄ and 50% CO₂ (red squares), (iii) 44.5% C₃H₂F₄ and 44.5% CO₂ (blue triangles).

First, efficiency and streamer probability have been measured for tetrafluoropropene based gas mixtures, with the addition of different concentrations of CO₂. In figure 5 mixtures with different C₃H₂F₄ and CO₂ ratio are shown, while s-C₄H₁₀ and SF₆ are kept constant. By increasing C₃H₂F₄ and decreasing CO₂ the WP is shifted towards higher voltages, while there is no significant variation of the streamer probability at the WP.

In order to investigate the effects of the isobutane, we have also measured efficiency and streamer probability for tetrafluoropropene-based gas mixtures, with the addition of different concentrations of isobutane. As shown in figure 6 we can see a similar behaviour as for CO₂, leading to the conclusion that there is a strong dependence between the concentration of C₃H₂F₄ and the WP, due to the higher electron affinity of tetrafluoropropene with respect to tetrafluoroethane.

Fig. 6. Efficiency curves and streamer probability for gas mixtures with different ratios between C₃H₂F₄ and s-C₄H₁₀ (CO₂ and SF₆ are kept constant at 50% and 1% respectively): (i) 29% C₃H₂F₄ and 20% s-C₄H₁₀ (black dots), (ii) 39% C₃H₂F₄ and 10% s-C₄H₁₀ (red squares).
Since isobutane affects the gas mixture flammability, its reduction or, preferably, removal is desirable for safety reasons. In figures 7 and 8 the efficiency curves and the streamer probability for gas mixtures with different CO$_2$ and i-C$_4$H$_{10}$ ratio, with constant C$_3$H$_2$F$_4$ (34%) and SF$_6$ (1%), are shown. The WP does not vary monotonically with the ratio, decreasing by 100 V when raising the isobutane fraction from 0% to 5% and increasing by 500 V when the isobutane fraction is 20%, while there are very similar streamer probability in all cases (note that Fig. 8 refers to a shift between the applied HV and the HV at the 90% of efficiency). Although the reduction of isobutane is desirable, its reduction also results in a less steep turn-on of the efficiency curve.

![Efficiency curves](image7.png)

**Fig. 7.** Efficiency curves for gas mixtures with different ratios between CO$_2$ and i-C$_4$H$_{10}$ (C$_3$H$_2$F$_4$ and SF$_6$ are kept constant at 34% and 1% respectively): (i) 65.5% CO$_2$ and 0% i-C$_4$H$_{10}$ (black dots), (ii) 60.5% CO$_2$ and 5% i-C$_4$H$_{10}$ (red squares), (iii) 55.5% CO$_2$ and 10% i-C$_4$H$_{10}$ (blue triangles), (iv) 50% CO$_2$ and 15% i-C$_4$H$_{10}$ (green reverse triangles), (v) 44.5% CO$_2$ and 20% i-C$_4$H$_{10}$ (orange stars).

![Streamer probability](image8.png)

**Fig. 8.** Streamer probability for gas mixtures with different ratios between CO$_2$ and i-C$_4$H$_{10}$ (C$_3$H$_2$F$_4$ and SF$_6$ are kept constant at 34% and 1% respectively): (i) 65.5% CO$_2$ and 0% i-C$_4$H$_{10}$ (black dots), (ii) 60.5% CO$_2$ and 5% i-C$_4$H$_{10}$ (red squares), (iii) 55.5% CO$_2$ and 10% i-C$_4$H$_{10}$ (blue triangles), (iv) 50% CO$_2$ and 15% i-C$_4$H$_{10}$ (green reverse triangles), (v) 44.5% CO$_2$ and 20% i-C$_4$H$_{10}$ (orange stars).
The role of the SF$_6$ in the gas mixture is to suppress the streamers$^{[2]}$. Given this, we have studied tetrafluoropropane-based gas mixtures with different percentage of SF$_6$ and we have compared them with the ALICE standard gas mixture. In figures 9 and 10 the efficiency curves and the streamer probability for these gas mixtures are shown. A small variation of SF$_6$, from 0.3% to 1.0%, leads to a variation of the WP of $\sim$ 500 V, while there is no significant variation in the streamer probability when increasing SF$_6$ from 0.3% to 0.6%. The suppression of the streamers is slightly higher with 1.0% of SF$_6$.

Fig. 9. Efficiency curves for gas mixtures with different percentage of SF$_6$, compared with ALICE standard gas mixture (i) 0.3% SF$_6$, 89.7% C$_2$H$_2$F$_4$, 10% i-C$_4$H$_{10}$ (black dots). (ii) 0.3% SF$_6$, 39.7% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$ (red squares). (iii) 0.6% SF$_6$, 39.4% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$ (blue triangles). (iv) 1.0% SF$_6$, 39.3% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$ (green reverse triangles).

Fig. 10. Streamer probability for gas mixtures with different percentage of SF$_6$, compared with ALICE standard gas mixture (i) 0.3% SF$_6$, 89.7% C$_2$H$_2$F$_4$, 10% i-C$_4$H$_{10}$ (black dots). (ii) 0.3% SF$_6$, 39.7% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$ (red squares). (iii) 0.6% SF$_6$, 39.4% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$ (blue triangles), (iv) 1.0% SF$_6$, 39.3% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$ (green rhombuses).
3.1. Most promising gas mixtures (up to now)

Given the results shown above, we concluded by defining two promising gas mixtures. In figures 11 and 12 the efficiency curves and the streamers probability for these two gas mixtures, compared with the ALICE standard mixture, are shown.

The one with 39.7% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$, 0.3% SF$_6$ has a GWP = 72 (  ~ 20 times lower than the ALICE mixture), but the WP is ~ 1 kV higher. The streamer probability is also higher than that with the ALICE mixture.

The one with 39.0% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$, 1.0% SF$_6$ has a GWP = 232 (  ~ 5 times lower than the ALICE mixture) and a WP ~ 1.5 kV higher than the standard mixture. The streamer probability is similar to that with the ALICE mixture, because of the higher percentage of SF$_6$. 

![Fig. 11. Efficiency curves for the two most promising gas mixtures (up to now) compared with the ALICE standard gas mixture (i) (black dots). (ii) 39.7% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$, 0.3% SF$_6$ (red squares), (iii) 39.0% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$, 1.0% SF$_6$ (blue triangles).](image1)

![Fig. 12. Streamer probability for the two most promising gas mixtures (up to now) compared with the ALICE standard gas mixture (i) (black dots). (ii) 39.7% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$, 0.3% SF$_6$ (red squares), (iii) 39.0% C$_3$H$_2$F$_4$, 50% CO$_2$, 10% i-C$_4$H$_{10}$, 1.0% SF$_6$ (blue triangles).](image2)
4. R&D at Gamma Irradiation Facility (GIF) at CERN

4.1. Gamma Irradiation Facility (GIF++)

Finding a new eco-friendly gas mixtures with suitable performance is not enough. It is also important to perform an ageing test on the new gas mixtures in order to see if they allow for stable long-term operations. Ageing is strongly influenced by the chemical composition of the gas mixture: in the ALICE MID RPCs ageing is mainly due to a deterioration of the electrodes surface smoothness probably due to UV and chemical action. This causes the formation of tips and leads to an increase of the dark current\(^{18}\).

For this reason we tested RPCs under gamma irradiation to simulate many years of operations at the LHC while keeping track of current drawn over time. These tests are performed at the CERN Gamma Irradiation Facility (GIF++). ECO-gas@GIF++ collaboration (ALICE, ATLAS, CMS, EP-DT, SHiP) has been created to perform these aging tests on HFO-based gas mixture with low GWP. RPCs have been provided by the different groups and have been installed onto a common trolley placed inside the bunker at GIF++. The GIF is equipped with a Cs137 radioactive source (14 TBq), emitting 662 keV gamma rays. Filters are used to modulate the radiation on the detectors under test and there is the possibility to have muon beam for beam tests.

4.2. Ageing test

The ageing test started in 2019, with two mixtures called ECO1 and ECO2. ECO1 is constituted by CO\(_2\), HFO, i-C\(_4\)H\(_{10}\), SF\(_6\) in the proportion 50/45/4/1. ECO2 is constituted by the same gases in the proportion 60/35/4/1. The low percentage of isobutane allows to treat these mixture as non-flammable.

The chambers inside the GIF bunker are kept at a HV value close to the WP of the RPC and are irradiated for a prolonged amount of time (stability test). Every week a high voltage scan is performed without source in order to measure the dark current (i.e. current with no source) and observe its long term behavior.

Dark current as a function of HV with the ECO1 gas mixture for the ALICE RPC are shown in the plots in figures 13 and 14 respectively before and after the stability test (5 months between the two tests). We can observe an increase of the dark current. The fit used to estimate the Ohmic component of the dark current at 11.4 kV is shown in red.
Fig. 13. Dark current drawn by the ALICE RPC as a function of the HV before the start of the irradiation, while flushed with ECO1 mixture.

Fig. 14. Dark current drawn by the ALICE RPC as a function of the HV after the integration of 22 mC/cm$^2$, while flushed with ECO1 mixture.

In figures 15 and 16 the dark current vs. HV for the ECO2 gas mixture for the ALICE RPC, before and after the stability test, are shown. By increasing the percentage of CO$_2$, we have observed a large increase of the current as soon as we started the tests with ECO2 and the reasons for this increase are still under investigation (see Fig. 16). After that we had to remove the chamber since it was damaged: tests have continued with other RPCs from the collaboration and results for ECO2 are under investigation.
Fig. 15. Dark current drawn by the ALICE RPC as a function of the HV before the start of the irradiation, while flushed with ECO2 mixture.

Fig. 16. Dark current drawn by the ALICE RPC as a function of the HV after few days of short irradiation period, while flushed with ECO2 mixture.

In the plots in figures [17 and 18] the Ohmic component (extrapolated from low HV linear fit from I-V curve) of the current for both the ECO1 and ECO2 gas mixture, at the RPC working point, is shown. Each point in the plots corresponds to an HV scan performed once a week, without source. The current increases over time for reasons still under investigation.
5. Conclusions

Following R&D studies on low GWP gas mixtures, C\textsubscript{3}H\textsubscript{2}F\textsubscript{4} seems a possible candidate to replace C\textsubscript{2}H\textsubscript{2}F\textsubscript{4}. However, the direct replacement of tetrafluoroethane with tetrafluoropropane is not possible because of the high WP (\textgreater 14 kV). It has then been decided to add CO\textsubscript{2} to the gas mixture, in order to operate at lower voltages. Promising gas mixtures consisting of C\textsubscript{3}H\textsubscript{2}F\textsubscript{4}, CO\textsubscript{2}, \textit{i}-C\textsubscript{4}H\textsubscript{10} and SF\textsubscript{6}, allow for a factor 5-20 GWP reduction with respect to the standard gas mixture, depending on the percentage variation of the different gases.

At GIF++ we have observed an increase of both ohmic and working current with the ECO gas mixtures under test; the link with CO\textsubscript{2} percentage in the gas mixture is under investigation. Tests beams have been performed in July, September and October 2021 at GIF++ with other RPC groups, in order to do more tests on ECO2 gas mixture, and these results are under analysis.
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