Characterization of RPC operation with new environmental friendly mixtures for LHC application and beyond

2016 JINST 11 C07016
(http://iopscience.iop.org/1748-0221/11/07/C07016)

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Characterization of RPC operation with new environmental friendly mixtures for LHC application and beyond

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Abstract: The large muon trigger systems based on Resistive Plate Chambers (RPC) at the LHC experiments are currently operated with R134a based mixture. Unfortunately R134a is considered a greenhouse gas with high impact on the environment and therefore will be subject to regulations aiming in strongly reducing the available quantity on the market. The immediate effects might be instability on the price and uncertainty in the product availability. Alternative gases (HFO-1234yf and HFO-1234ze) have been already identified by industry for specific applications as replacement of R134a. Moreover, HFCs similar to the R134a but with lower global warming potential (GWP) are already available (HFC-245fa, HFC-32, HFC-152a). The present contribution describes the results obtained with RPCs operated with new environmental friendly gases. A particular attention has been addressed to the possibility of maintaining the current operation conditions (i.e. currently used applied voltage and front-end electronics) in order to be able to use a new mixture for RPC systems even where the common infrastructure (i.e. high voltage and detector components) cannot be replaced for operation at higher applied voltages.

Keywords: Gas systems and purification; Gaseous detectors; Materials for gaseous detectors; Resistive-plate chambers
1 Introduction

Resistive Plate Chambers (RPCs) [1] are gaseous parallel-plate detectors that combine high time resolution (∼ 1 ns) with good spatial resolution (∼ 1 cm). RPCs are extensively employed at the Large Hadron Collider (LHC) experiments as a part of their muon trigger systems. They are used to identify unambiguously the relevant bunch crossing at which the muon tracks are associated, even in presence of the high rate and background expected (up to 1000 Hz/cm²).

Nowadays, RPCs are suitably operated with a three component non-flammable gas mixture: 94.7 % of R134a (C₂H₂F₄), 5 % of iC₄H₁₀ and 0.3 % of SF₆ [2]. Water vapour is added to the gas mixture in order to maintain a relative humidity of about 45 % (i.e. 8000–12000 ppmV) and avoid any changes of the High Pressure Laminates (HPLs) resistivity. Due to the large detector volume (about 16 m³ per each experiment) and the use of a relatively expensive gas mixture RPC are already operated with gas recirculation systems [3]. Unfortunately, R134a and SF₆ are not only expensive but also they are considered greenhouse gases (GHG).¹ Their production and use is subject to a new European Union (EU) regulation starting from January 2015 [4]. The EU regulation on fluorinated gases is aiming in limiting the total amount of F-gases that can be sold in EU from 2015 onwards with a reduction to one-fifth from 2014 to 2030, banning the use of F-gases where less harmful alternative are available and preventing the emission from existing equipment by requiring adequate checks, maintenance and recovery of the gases at the end of the equipment’s life. The EU regulation might have a significant effect on availability and price, especially for R134a where other alternative

¹GHGs are gases which absorb and emit radiation within the thermal infrared range and therefore they are responsible for the greenhouse effect. GHGs are classified according to their Global Warming Potential (GWP) which is a relative measure of how much heat a GHG traps in the atmosphere with respect to the same mass of CO₂. GWPs for R134a and SF₆ are 1430 and 23900 respectively.
gases are already available for refrigeration systems. In a short-term scale, the SF$_6$ should be less affected since no valid alternative gases have been found yet.

Despite the use of gas recirculation systems, the RPC detector systems largely dominate the greenhouse gas emission from particle detection at CERN [5]. RPCs are responsible for about 80% of the total CO$_2$ equivalent emission. In fact, the recirculation rate is limited to about 86% by the presence of leaks at the detector level. However, even if we could imagine to repair all leaks, past tests [6] have validated the RPC operation at LHC only up to 95% gas recirculation. Therefore, further test will be needed to verify stable long-term operation at HL-LHC conditions with 95% or higher gas recirculation.

An alternative and long term approach is to explore the possibility to operate RPC detectors with new low GWP gases. Preliminary results of this research line will be described in the following.

2 RPC operation with new environmental friendly gases

2.1 Characterization of alternative gases for RPC operation

Several alternative gases with molecular structure similar to the R134a exist or have been recently developed and produced to substitute the R134a in industrial application.

Old alternative gases can be found in the Hydro-Fluoro-Carbon (HFC) group (the same group or R134a). These molecules are characterized by simple Carbon-Carbon bounds and by the presence of Fluorine and Hydrogen atoms. Examples of HFCs are the R245fa, R32 and R152a (GWPs are 1030, 675 and 120 respectively).

New recently developed gases are part of the Hydro-Fluoro-Olefine family (HFO) and they are characterized by the presence of a double Carbon-Carbon bound. HFOs are already used by air conditioning and refrigerator industry to replace the R134a. Examples of HFOs are the HFO-1234yf and HFO-1234ze (GWPs are 4 and 6 respectively).

Refrigerant properties of HFCs and HFOs are very well know while studies of ionization process in particle detectors are just started. The main issues for HFCs and HFOs use with particle detectors are related to the flammability of many of these low GWP gases, to their low vapour pressure at ambient temperature and to the overall purity grade of the available gases. The flammability represents a problem because of the presence of leaks in the current RPC systems. In addition, the use of flammable mixtures requires the installation of flammable gas detection close to the detector which is increasing the complexity of the system. The low vapour pressure (below 1 barg) represents a limitation to the maximum flow and it might lead to vapour condensation in the devices used to control the gas flow of each mixture component, i.e. gas mass-flow controllers (MFCs). The formation of liquid in the MFCs or the deposition of oily impurities are responsible of drifts in the MFCs calibration causing wrong mixture composition. Gas chromatographic analyses have been performed for each gas mixture tested. The corresponding gas chromatograms are shown in figure 1. The overall quality is good (impurities concentration is below few hundreds ppmV). The main impurity is always air with the exception of R245fa and HFO-1234ze where further investigations are needed.

The last remaining RPC open gas system (ALICE — RPC muon trigger) has been converted to gas recirculation during the LHC long shutdown 1, i.e. 2013–2014.
Figure 1. Gas chromatograms of the new environmental HFCs and HFOs used in the present research.

2.2 Experimental setup

Figure 2 shows a schematic view of the experimental set-up. Two standard HPL RPCs with gas gap 2 mm wide and a surface of $80 \times 100$ cm$^2$ were used. Two scintillators were employed to select cosmic muons intersecting the two RPCs under test. The RPCs active area selected corresponds to seven strips (each strip is 2 cm wide). Data were acquired using a CAEN Waveform Digitizer VME V1730 [7]. Several multi-components gas mixtures have been prepared using a flexible gas mixing unit able to control up to 6 different gases. For the new gases, the calibration data for Bronkhorst thermic mass-flow controllers have been measured with a dedicated setup [8]. The gas mixtures were monitored with a GC-MS during all tests.

Figure 2. Schematic view of the experimental setup used for testing environmental friendly gas mixtures for the RPC detectors.

A complete scan of the efficiency curve has been done for each mixture. The raw data extracted with the CAEN Digitizer consists of the time evolution of the pulse for each strip. The pulses are
then processed to obtain pulse charge distribution, pulse height distribution, detector efficiency, pulse charge as a function of the applied voltage (or efficiency) and cluster size as a function of the applied voltage (or efficiency). Figure 3 and figure 4 show an example of the plots used for the analysis in the case of R32 based mixtures. The detector was considered efficient if the induced pulse height on the read-out strip was greater than 0.5 mV and its total charge was larger than 100 fC. Signals with induced pulse charge higher than 3 pC have been considered as streamers.

![Pulse height distribution](image1.png)  
![Pulse charge distribution](image2.png)  

**Figure 3.** RPC pulse height and charge distributions for R32 based mixture at different applied high voltage.

![Efficiency/streamer](image3.png)  
![Pulse charge vs HV](image4.png)  
![Cluster size](image5.png)  

**Figure 4.** RPC detector efficiency, pulse charge and cluster size as a function of the applied voltage for R32 based mixture.

### 3 Experimental results

In this section a detailed overview of the results obtained is given. More than 50 different mixtures have been tested with 6 different HFCs and HFOs. Summary and comparison of the results will be presented in the next section.

#### 3.1 HFO-1234yf and HFO-1234ze based mixtures

HFO-1234yf and HFO-1234ze are two isomers of Tetrafluoropropene (C₃H₂F₄). HFO-1234yf is performing better as a refrigerant fluid but it is classified as slightly flammable, while HFO-1234ze is not flammable.

Since RPC detectors operated with HFO-1234yf and HFO-1234ze based mixtures have shown very similar performances, most of the tests have been performed with HFO-1234ze. Moreover, given the similarity between the two HFOs, in the following we will refer simply to HFO-1234.
The first part of the test was aiming in understanding if HFO-1234 can replace one of the current RPC mixture components: R134a, SF$_6$ or iC$_4$H$_{10}$. HFO-1234 revealed not to be a strong electronegative gas, i.e. it cannot substitute the SF$_6$. Moreover, when HFO-1234 is used directly in place of R134a, RPC detector are not efficient below 15 kV [9]. In order to move the detector efficiency range towards lower applied voltage values (compatible with the current high voltage infrastructure used at the LHC experiments), Argon or Helium were added at different moments. Figure 5 shows the results obtained by adding Argon with respect to pure HFO-1234 and standard R134a based mixture (in the following also indicated as ATL-CMS mixture, since it is used by the ATLAS and CMS RPC muon trigger systems). About 40% of Argon is needed to obtain a working voltage of about 9 kV (which is about the standard value for RPC operated with R134a based mixture). The GWP equivalent of these HFO-1234-Argon mixtures is about 150 but a large fraction of streamer events is present, i.e. about 50% streamers at 50% efficiency, which is well above the maximum tolerable limit for operation at the LHC experiments (i.e. below 5% at full efficiency).

![Figure 5. Comparison of the RPC detector efficiency and streamer probability for HFO-1234 based mixture, HFO-1234-Argon based mixture and standard R134a based mixture.](image)

The addition of Helium instead of Argon gives better results (figure 6): RPC detectors are efficient at about 9 kV, the fraction of streamer is significantly lower but still there is not an operation region streamer-free.

![Figure 6. Comparison of the RPC detector efficiency and streamer probability for HFO-1234-Helium based mixture and standard R134a based mixture.](image)
Good results have been obtained with mixtures where the R134a was only partially replaced by HFO-1234 (figure 7). In particular, the five component mixture HFO-1234 37.5% - R134a 37.5% - iC₄H₁₀ 4.5% - SF₆ 0.6% - He 20% shows an efficiency plateau without streamers almost equivalent to the standard RPC R134a based mixture. Increasing the HFO-1234 concentration with consequent reduction of R134a produces a reduction of the operation region without streamers.

Figure 7. Comparison of the RPC detector efficiency and streamer probability for HFO-1234-R134a based mixture and standard R134a based mixture.

3.2 R32 based mixtures

R32 (or HFC-32) is a Difluoromethane. It is a HFC with very simple chemical structure. Its GWP is 675. Figure 8 shows the results obtained. The working point is at about 7–8 kV but the streamer probability increases very rapidly and given the large cluster size measured (figure 4) a poor quenching capacity can be inferred.

Figure 8. Comparison of the RPC detector efficiency and streamer probability for HFC-32 based mixture and standard R134a based mixture.

3.3 R152a based mixtures

R152a (or HFC-152a) is a 1,1-Difluoroethane. The chemical structure is equal to the R134a, i.e. ethane. However, HFC-152a has half of the Fluorine atoms with respect to R134a per molecule. Figure 9 shows efficiency curves and streamer probability for HFC-152a based mixtures. As
expected, given the similarity in the molecular structure, the RPC detectors are efficient at about the same voltage (i.e. 9.5 kV). However, probably due to the lower number of Fluorine, HFC-152a is less effective than R134a in suppressing the streamers and therefore a higher SF6 concentration was needed in order to obtain a similar operation region streamer-free.

In addition, for potential future operation of large size RPC particle detectors, it has to be considered that HFC-152a is flammable.

3.4 R45fa based mixtures

HFC-245fa (or R245fa) is a 1,1,1,3,3-Pentafluoropropane. Therefore the basic structure is based on the propane chain (three Carbon atoms with only simple bonds). For this gas it was not possible to operate efficiently RPC detector below 11 kV (figure 10). The addition of 50% Helium allowed to move the efficiency curve to about 7 kV. However, the operation region with low streamer probability is much narrow with respect to the R134a based mixture.
4 Conclusions

An intense R&D activity focused on alternative mixtures for RPC detectors has been started in many laboratories [10] for two main reasons. The current mixture recirculation rate for the ATLAS and CMS RPC systems cannot be increased due to the presence of leaks at the detector level very difficult to repair. Moreover, there is a strong interest in moving towards new eco-friendly gases with low GWP (below 500). In fact, R134a and SF6 consumption are subject to a new EU regulation aiming in limiting the total amount of high GWP gases that can be sold in the EU. The objective is an overall reduction to one-fifth from 2014 to 2030. Table 1 summarizes the results obtained using as key parameters the applied high voltage at 50% efficiency (HV), the streamer probability at 50% efficiency (Stream), the pulse charge (for both avalanche and streamer), the voltage difference between 50% efficiency and 50% streamer probability (ΔV Eff-Stream) and the cluster size (Clu.size). More than 50 mixtures have been tested with 6 different HFCs and HFOs.

<table>
<thead>
<tr>
<th>Mixture</th>
<th>Chem. Struct.</th>
<th>GWPmix</th>
<th>HV (V)</th>
<th>Stream (%)</th>
<th>Pulse charge (pC)</th>
<th>ΔV Eff-Stream (V)</th>
<th>Clu.size (strip)</th>
</tr>
</thead>
<tbody>
<tr>
<td>R134a - iC4H10 4.5 - SF6 0.3</td>
<td>C-C</td>
<td>1491</td>
<td>9581</td>
<td>1.5</td>
<td>0.5 / 6.0</td>
<td>1000</td>
<td>1.5</td>
</tr>
<tr>
<td>R134a - iC4H10 4.5 - SF6 0.3</td>
<td>C-C</td>
<td>1263</td>
<td>6666</td>
<td>2.0</td>
<td>1.0 / 7.0</td>
<td>610</td>
<td>2</td>
</tr>
<tr>
<td>HFO1234 - iC4H10 4.5 - SF6 0.3 - Ar 42.5</td>
<td>C-C-C</td>
<td>134</td>
<td>8790</td>
<td>7.0</td>
<td>2.0 / 15.0</td>
<td>160</td>
<td>4</td>
</tr>
<tr>
<td>HFO1234 - iC4H10 4.5 - SF6 0.6 - He 50</td>
<td>C-C-C</td>
<td>373</td>
<td>9020</td>
<td>22.0</td>
<td>1.5 / 8.0</td>
<td>700</td>
<td>4</td>
</tr>
<tr>
<td>HPO1234 - iC4H10 4.5 - SF6 0.6 - He 20</td>
<td>C-C-C</td>
<td>889</td>
<td>10450</td>
<td>1.8</td>
<td>0.5 / 5.8</td>
<td>970</td>
<td>1.6</td>
</tr>
<tr>
<td>HPO1234 - iC4H10 4.5 - SF6 0.6 - He 20</td>
<td>C-C-C</td>
<td>726</td>
<td>10500</td>
<td>8.0</td>
<td>0.5 / 6.5</td>
<td>700</td>
<td>1.6</td>
</tr>
<tr>
<td>HPO1234 - iC4H10 4.5 - He 20</td>
<td>C-C-C</td>
<td>434</td>
<td>10800</td>
<td>50.0</td>
<td>1.5 / 8.0</td>
<td>415</td>
<td>2.5</td>
</tr>
</tbody>
</table>

It has been observed that methane (C) and ethane (C2) molecular structures allow direct operation at applied high voltages similar to the ones currently used at the LHC experiments. On the contrary propane or propene structures (C3 without or with double bounds) require the addition of Argon or Helium. Argon is very easy to ionize and it has no quenching capacity while Helium does not participate to the ionization process and it is simply producing an effect similar to operation at a reduced effective pressure. Unfortunately mixtures with Argon and Helium show the presence of a large fraction of streamers well above the tolerable limit for safe and long term operation at the LHC experiments.

Encouraging results have been obtained with a partial (50%) substitution of the R134a with HFO-1234 and the addition of Helium (20%). As it can be seen from table 1, RPC detector performances with this mixture are very similar to the standard R134a based mixture in all the parameters used during the tests. The GWP of this HFO-1234 - R134a based mixture is 40% less with respect to the standard R134a mixture. However, further tests are needed to fine tune the mixture composition and there might be concerns for using large quantity of Helium in the current LHC experiments due to the presence of photomultipliers.

For future RPC applications where the high voltage system and the detector can be designed for much higher applied voltage (about 15 kV or higher for 2 mm gas gap) or lower threshold front-end electronics, it is interesting to explore operation with only HFO-1234 based mixture.
References


