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# Towards more eco-friendly gaseous detectors

M.C. Arena [b], R. Guida [a], S. Juks [d], B. Mandelli [a,*], G. Rigoletti [a], M. Verzeroli [c]

[a] CERN, Meyrin, Switzerland
[b] University of Pavia, Pavia, Italy
[c] University of Lyon, Lyon, France
[d] University of Saclay, Saclay, France

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ABSTRACT

In the last years, the particle detector community faced a new challenge: how to convert existing and future gaseous detectors in more eco-friendly ones. Indeed, several detectors make use of greenhouse gases (GHGs) since they allow achieving excellent performance and long-term stability. With a growing concern on climate change and future restrictions, it is fundamental to look for solutions that can balance detector performance with an eco-friendly approach.

CERN was a pioneer in developing strategies to reduce the use of GHGs in particle detection. Three different strategies have been implemented: the use of gas recirculation and recuperation systems for existing and future detector systems and the search of alternative eco-friendly gas mixtures.

Thanks to the first two approaches, it is possible to recycle the gas mixture supplied to the detectors and to retrieve GHGs from the used gas mixtures, allowing a reduction of GHG emissions up to 95%–100% at the LHC experiments. By looking at the long-term operation and future particle detector applications, the search of alternative eco-friendly gas mixtures must be envisaged. A big effort is on-going for the $C_2H_2F_4$ replacement for the Resistive Plate Chamber (RPC) detectors, which nowadays account for most of CERN particle detector emissions. Several eco-friendly gas mixtures have been identified and tested but finding a suitable replacement for the LHC experiments is particularly challenging. Alternatives to $SF_6$, which is the most powerful GHG, are under studies for RPCs in term of detector performance and chemical characterisation. Last point is increasingly crucial since most of the so-called eco-friendly gases belong to the PFAS family which is very likely to be subject to new regulations soon.

## 1. Introduction

At CERN, greenhouse gas (GHG) emissions arise from the operation of the Laboratory's research facilities. The majority of these emissions comes from CERN's core experiments and more than 78% are fluorinated gases used for particle detection and detector cooling for a total of about 150000 $tCO_2e$ in 2022 [1]. In particular, for particle detection, the F-gases employed are $C_2H_2F_4$, $SF_6$, $CF_4$ and $C_4F_{10}$. In Run 2, the $C_2H_2F_4$ and $SF_6$ contributed for more than 80% of particle detection emissions due to their usage in the ATLAS and CMS Resistive Plate Chamber (RPC) systems where several leaks are present at the detector level. The CERN's objective is to reduce its scope emissions by 28% by the end of Run 3 (with respect to 2018 data). To fulfil this objective and to further pursuit the GHG reduction, the CERN EP-DT Gas Team, the CERN Environmental Protection Steering board (CEPS) and the LHC experiments elaborated a strategy based on three action lines, which will be described in the following sections [2]:

**Gas Recirculation.** The gas mixture is taken at the output of the detectors, purified and sent back to the detectors. It is technically possible to recycle 100% of the gas mixture.

**Gas Recuperation.** The gas mixture is sent to a recuperation plant where the GHG is extracted, stored and re-used.

**Alternative gases.** Search of alternative gas mixtures suitable for particle detectors that do not contain or limit the use of GHGs.

The reduction of the use of F-gases is fundamental for next LHC Runs and future particle detector applications also because of the implementation in Europe of the F-gas regulation [3]. This regulation, recently updated, establishes the total elimination of hydrofluorocarbons by 2050. A phasing down policy for the reduction of national quota of F-gas refrigerants is in progress and the F-gas availability in 2030 (LHC Run 4) will be less than 25% with respect to 2014 data. This has already an impact on prices [4], which are increasing, obviously leading to an increase in the operational cost of detector systems.

* Corresponding author.
*E-mail address:* beatrice.mandelli@cern.ch (B. Mandelli).

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Furthermore the availability of these gases for the time to come is not known and this aspect has to be taken into account seriously for future detector operation. It has also to be mentioned that in 2023 the European Chemicals Agency (ECHA) released a proposal regarding restriction on PFAS, *i.e.* per- and polyfluoroalkyl substances, which contains at least one fully fluorinated methyl (CF3-) or methylene (-CF2-) carbon atom (without any H/Cl/Br/I attached to it). The proposal envisages to cover over 10,000 different PFAS, which are considered environmental pollutants with links to harmful health effects. Most of the so-called "eco-friendly" gases belong to the PFAS family.

## 2. Gas recirculation and recuperation systems

The gas recirculation systems are the first way to reduce GHG emissions for now and for future detectors. In general, it is technically possible to recycle 100% of the gas mixture, *i.e.* reduce emissions almost to zero. These recirculation gas systems are complex apparatus that have to ensure an extremely high reliability in terms of stability and quality of the gas mixture delivered to the detectors. In particular possible pressure and flow fluctuations in the system as well as creation of impurities have to be addressed [5].

In some cases, it is not possible to recirculate 100% of the gas mixture due to detector constrains. In these cases, a fraction of gas mixture is replenished with fresh one and it is exhausted to the atmosphere. Instead of venting it to the atmosphere, this fraction of gas mixture can be sent to a recuperation plant where the most valuable component is extracted, stored and then re-used. Recuperation of a single gas component from a gas mixture is often challenging both in term of recuperation efficiency and quality of recuperated gas. The recuperation plants are custom-made systems specifically built for the detector gas mixtures. At CERN several gas recuperation systems are employed for GHGs and expensive gases, as the ones for CMS-CSC ($CF_4$), ATLAS-TGC ($nC_5H_{12}$), ALICE TRD (Xenon), ATLAS TRT (Xenon), LHCb-RICH1 ($C_4F_{10}$), LHCb-RICH2 ($CF_4$) and CMS RPC ($C_2H_2F_4$). Thanks to these gas recuperation systems the GHG emissions can be further reduced.

## 3. Alternative gases

In the last years, industry developed several new gases and liquids as alternatives to GHGs to meet the challenges presented by the EU regulations. These new gases have been developed for industrial applications as refrigerants or high voltage insulating medium, so their usage in gaseous detector is not straightforward as the gas properties may not be adequate. The industrial replacements of R134a are the HydroFluoroOlefyns (HFOs), which have a GWP less than 6. Several HFOs are available on the market and the HFO1234ze has been selected as possible replacement for the RPC detectors. Concerning $SF_6$, possible industrial alternatives with low GWP are the HFO12224yd and 3M Novec 5110 ($CF_3C(O)CF(CF_3)_2$) and 3M Novec 4710 ($(CF_3)_2CFCN$).[1] The last two gases belong to the PFAS family. It is important to consider that two factors identify the greenhouse gases and their effects on climate: the radiative efficiency and lifetime in the atmosphere. The lower are the GWP and the lifetime, the easier is the creation of sub-products in the atmosphere. The three factors that determine the atmospheric lifetime are: rain out (water solubility), oxidation (reactivity with OH) and photolysis (UV absorbance). These three mechanisms are present in several gaseous detectors and they have to be taken into account for the detector operation when eco-friendly gases are used. For example the Novec 5110 has a strong absorbance in the near UV while the Novec 4710 can react with water creating an amide in a concentration of few ppb.

[1] It has in any case to be considered that 3M company will stop to produce PFAS by 2025. Other companies will continue the production.

## 4. Alternatives gas mixtures for the RPC detectors

Given the large impact of RPCs in the GHG emissions at CERN, a particular effort has been invested to search for new eco-friendly gas mixtures for the RPC detectors. Their gas mixture is made of ∼95% $C_2H_2F_4$, 4.5% $iC_4H_{10}$ and 0.3% $SF_6$ with 40% relative humidity.

For the replacement of $C_2H_2F_4$ two research lines have been investigated: a mid-term solution with the partial replacement of the $C_2H_2F_4$ with $CO_2$ and studies on the use of HFO1234ze. Studies are also on-going to replace the $SF_6$ as it will be discussed in Section 4.2.

The studies of new eco-friendly gas mixtures are performed in the laboratory for a complete characterisation of the detector as well as at the CERN Gamma Irradiation Facility (GIF++) to evaluate long-term detector performance and possible ageing effects. The operation of RPC with these gases is not straightforward, especially for the LHC RPC systems where it is not possible to replace the detectors themselves as well as most of the infrastructures (*i.e.* high voltage systems, cables, front-end electronics). The RPCs tested are High Pressure Laminate RPCs with a 2 mm gas gap and read-out strips 2.1 cm wide. A dedicated gas system unit is used to prepare the gas mixture with up to 6 gas components. The data acquisition is performed with a CAEN Digitizer V1730 without any amplification module. In this way it is possible to analyse each waveform in term of pulse height, charge and time in order to characterise the detector signals. The RPC performance are then evaluated by measuring efficiency, streamer probability, rate capability, induced charge for avalanche and streamer signals, cluster size and time resolution.

### 4.1. $C_2H_2F_4$ replacements

The mitigation of the GHG emissions from LHC RPC systems can be achieved simply by partially replacing the $C_2H_2F_4$ with the addition of an inert (or almost inert) gas. $CO_2$ has been selected as best compromise [6]. The addition of $CO_2$ decreases the operational voltage and it improves the time resolution, but it increases the streamer probability and it broadens the charge distribution, as it is visible in Fig. 1. To keep a low streamer probability the $SF_6$ concentration has to be increased up to 1%. The $CO_2$-based gas mixture with 1% $SF_6$ has also higher current (>15%) under irradiation with respect to the standard gas mixture. Long-term performance studies are therefore necessary to evaluate any possible effect. The gas mixture 30% $CO_2$, 64% $C_2H_2F_4$, 5% $iC_4H_{10}$ and 1% $SF_6$ has been selected by the ATLAS experiment and it is used since summer 2023 in the RPC system. This change of gas mixture will imply a reduction of ∼15% of GHG emissions and a cost saving of ∼ 100.000 CHF per year.

If the $C_2H_2F_4$ is replaced with the HFO1234ze, a too high applied voltage is necessary to reach the plateau efficiency in the LHC RPC of 2 mm gap. To overcome this problem, $CO_2$ is added to lower the working point. Also in this case, it is necessary to increase the $SF_6$ concentration to 1% to keep the streamer probability to an acceptable level. Fig. 2 reports, as an example, the efficiency curves and streamer probability of three eco-friendly gas mixtures. The gas mixture containing 45% HFO1234ze has a too high working point for the operation in LHC experiments. If the HFO1234ze is decreased to 25% the mean charge distribution is higher and the streamer probability is not negligible. This implies also an increase of currents. To overcome these limitations, a gas mixture containing 50% $CO_2$ and 22% $C_2H_2F_4$ and 22% HFO1234ze has been tested. In this case the working point is acceptable and the streamer probability is similar to the one of the standard gas mixture. Obviously the GWP of this gas mixture is higher with respect to the other two but it could be a possible compromise between detector performance and environment.

Given the usage of new gases, the RPC long-term operation with eco-friendly gas mixtures under high background radiation and possible ageing effects must be investigated. A long-term test is on-going at GIF++ lead by the EcoGas@GIF++ collaboration [7]. Several RPCs are

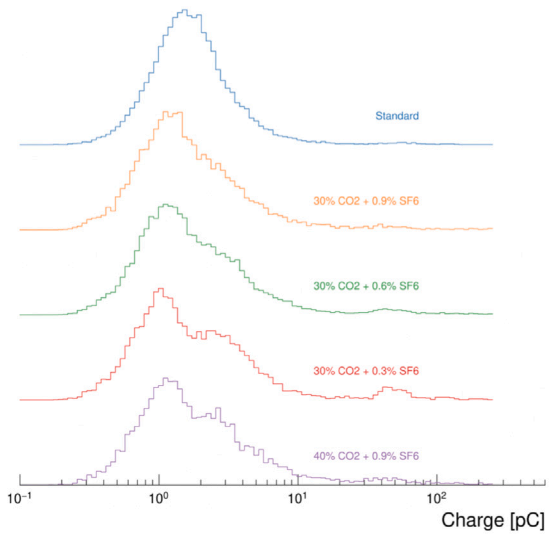


**Fig. 1.** Charge distribution for RPC standard gas mixture and gas mixture where 30% and 40% $CO_2$ is added (as replacement of $C_2H_2F_4$). In the case of 30% $CO_2$, the charge distribution is reported for different concentrations of $SF_6$. The $iC_4H_{10}$ concentration is kept at 5%.

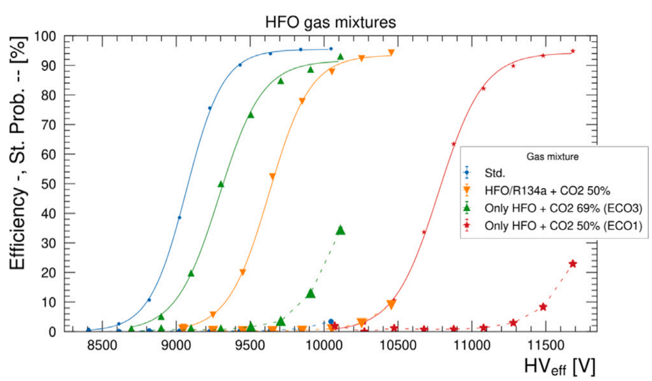


**Fig. 2.** Efficiency curves (continuous line) and streamer probability (dotted line) for the standard gas mixture and three eco-friendly gas mixtures. For the last, the $SF_6$ and $iC_4H_{10}$ concentrations are 1% and 4.5%, respectively.

under test from different experiments and detector performance have been studied for three gas mixtures containing different amount of HFO1234ze. The gas mixture 35% HFO1234ze, 60% $CO_2$, 4% $iC_4H_{10}$ and 1% $SF_6$ has been selected for long-term test. The currents under irradiation are constantly monitored and the dark and ohmic currents are measured every week. The detector performance are evaluated during test-beam periods. A total of ∼100 mC/cm$^2$ has been integrated up to now by all the detectors of the ECOgas collaboration and preliminary results show an increase in absorbed currents and a shift in the working point of around 200 V. Further analyses are ongoing to find any possible correlation.

### 4.2. $SF_6$ replacements

The $SF_6$ is used in a concentration of 0.3% in the RPC standard gas mixture but, given its very high GWP, it contributes for about ∼5% in the GWP of the RPC gas mixture. Furthermore in the updated EU F-gas regulation, the $SF_6$ is now included in the phasing-out policy. It is therefore important to study possible alternatives. In this study the $SF_6$ has been replaced in the standard RPC gas mixture with the different alternative gases in several concentrations (usually from 0.1% to 2%). Fig. 3 shows the streamer probability as a function of the

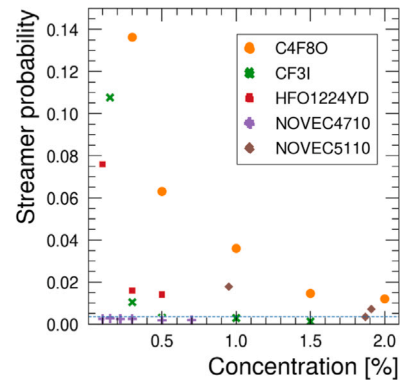


**Fig. 3.** Streamer probability as a function of the concentration for 5 different gases. The blu line represents the streamer probability value for the use of 0.3% $SF_6$ in the standard RPC gas mixture.

concentration for 5 different gases. Their GWP is <1 except for NOVEC 4710 (GWP 2100) and $C_4F_8O$ (GWP ∼8000). The use of $C_4F_8O$ has been discarded as a high concentration is necessary and therefore the gas mixture would have a higher GWP. The $CF_3I$ shows very good results but it does not satisfy CERN safety requirements as it is toxic. For the Novec 4710, a concentration of 0.1% is already enough to get the same streamer probability of 0.3% of $SF_6$. Also the Novec 5110 and the HFO12224yd shows good performance: it is enough 0.7% and 1.5% of them respectively to achieve a low streamer probability. The drawback of the Novec gases is that they could have reaction during detector operation: the Novec 4710 reacts with water[2] and the Novec 5110 is sensitive to UV radiation. Studies on long-term performance and possible chemical reaction have also to be conducted for the HFO1224yd as it contains an atom of Cl, which effects are not known in gas mixtures and detector operation.

## 5. Conclusions

With climate change a growing concern and implementation of F-gas regulations, it is fundamental for existing and future particle detector applications to reduce GHG emissions and search for eco-friendly has mixtures.

The first way to reduce GHG emissions is to make use of gas recirculation systems. In case of no constrains at detector level and no presence of leaks, it is technically possible to reach almost 100% recirculation, *i.e.* no emissions. If it is not possible to recycle 100% of the gas mixture, recuperation systems can be envisaged. They are complex and custom-made systems permitting to extract the GHG, store and re-use it.

For the future, it will be fundamental to looks for alternative gas mixtures to get rid of GHGs. The replacement of the GHG mixtures for current LHC gaseous detectors is quite challenging since the new eco-friendly gases have different gas properties with respect to the F-gases used today and it is not possible to replace the detector infrastructure (HV power supply, electronics, etc.). It will be also fundamental to understand the detector lifetime with the new eco-friendly gases.

Finally, for the long term, it is fundamental to keep in mind that, in the future, possible new restrictions could be set not only for the use of F-gases but also for PFAS, which include some of the nowadays called eco-friendly gases.

[2] For the High Pressure Laminate RPC 40% of relative humidity is added to the gas mixture to keep constant the electrode resistivity.

## Declaration of competing interest

The authors declare that they have no known competing financial interests or personal relationships that could have appeared to influence the work reported in this paper.

## References

[1] CERN Environment Report Vol. 3, 2023, http://dx.doi.org/10.25325/CERN-Environment-2023-003.
[2] R. Guida, B. Mandelli, R & D strategies for optimizing greenhouse gases usage in the LHC particle detection systems, Nucl. Instrum. Methods A 958 (2020) 162135.
[3] Regulation (EU) No 513/2024 of the European Parliament and of the council on fluorinated greenhouse gases and repealing regulation (EC).
[4] J. Kleinschmidt, B. Gschrey, S. Barrault, Briefing paper: HFC availability on the EU market, Oko-Recherche.
[5] M. Capeans, R. Guida, B. Mandelli, Gas systems for particle detectors at the LHC experiments: Overview and perspectives, SPPHY 212 (2018) 9196, http://dx.doi.org/10.1007/978-981-13-1313-419.
[6] R. Guida, B. Mandelli, G. Rigoletti, Performance studies of RPC detectors operated with $C_2H_2F_4$ and $CO_2$ gas mixtures, NIM A 1049 (2023) 168088, http://dx.doi.org/10.1016/j.nima.2023.168088.
[7] The RPC ECOGas@GIF++ collaboration, exploring eco-friendly gas mixtures for resistive plate chambers: A comprehensive study on performance and aging. http://dx.doi.org/10.48550/arXiv.2402.19395.