

#### Report on 2024 RPC EcoGas@GIF++ activities @ GIF++

Luca Quaglia<sup>1</sup> on behalf of the RPC ECOgas@GIF++ collaboration

<sup>1</sup>INFN Torino

8<sup>th</sup> GIF++ annual user meeting

03/12/2024

# Overview

- Currently employed gas mixture and environmental issues
  - $HFO+CO_2$  as a possible eco-friendly alternative
- Test set-up at the CERN GIF++
  - RPC EcoGas@GIF++ collaboration
- Aging studies HFO-based gas mixtures
- Performance evolution over time
- Conclusions and outlook

## The currently employed gas mixture

- RPC working parameters depend on the gas mixture employed
- The currently-used gas mixtures at the LHC grant the following properties:
  - 1) High density of primary ion-electron pairs
  - 2) Relevant quenching properties
    - $\rightarrow$  Ability of capturing recombination photons without further ionization
  - 3) Enough electron affinity to capture free electrons, reducing the spatial size of the discharge



# The need for a new RPC gas mixture

- All currently employed RPC gas mixtures contain different fractions of R134a (> 90%) and  $SF_{4}$  (< 1%)  $\rightarrow$  Fluorinated greenhouse gases (F-gases)
- New EU regulations [1] to reduce the impact of F-gases
  - $\rightarrow$  Phase down of the production and consumption of F-gases
  - $\rightarrow$  Ban of the gases if a more eco-friendly alternative is available
  - $\rightarrow$  Reduction of emissions from existing equipment





Emission of greenhouse gases from CERN - B. Mandelli VCI 2022

- RPC systems are the main consumer of F-gases at CERN
  - $\rightarrow$  Mainly due to leaks at detector level (leak fixing campaign started during LS2)
  - $\rightarrow$  Need to find a more eco-friendly gas mixture

# A possible solution

- First efforts of LHC RPC groups focused on R134a replacement
- Industrial use: from R134a to hydro-fluoro-olefine (HFO) family of gases
  - → Similar chemical structure as R134a but lower Global Warming Potential<sup>1</sup>
  - $\rightarrow$  Among all HFOs, HFO-1234yf and HFO-1234ze are currently used



- 1:1 replacement of R134a with HFO not possible
  - $\rightarrow$  Lower effective first Townsend coefficient
  - $\rightarrow\,$  Working voltage of the detectors moves to over 15 kV
- HFO has to be diluted with other gases
  - → Studies with cosmic muons by different LHC RPC groups [2-5]
  - $\rightarrow$  CO<sub>2</sub> found to be the most promising candidate for dilution
  - $\rightarrow$  In-depth studies on RPCs long-term behavior with eco-friendly alternatives needed

#### The RPC EcoGas@GIF++ collaboration

- Collaboration among ALICE, ATLAS, CMS, LHCb/SHiP and CERN EP-DT-FS group
- Each group provided <u>one detector prototype</u>
   → Installed on two mechanical support in the GIF++ bunker

Detector	# of gaps	Gap thickness (mm)	Electrode thickness (mm)	Gap area (cm²)
ALICE	1	2	2	2500
ATLAS	1	2	1.8	550
EP-DT	1	2	2	7000
CMS	2 (TW/TN + BOT)	2	2	3637 + 4215
LHCb/SHiP	1	1.6	1.6	7000
KODEL-H	2	1.4	1.43	2500

Name	R134a (%)	HFO (%)	CO <sub>2</sub> (%)	i-C <sub>4</sub> H <sub>10</sub> (%)	SF <sub>6</sub> (%)
STD (reference)	95.2	0	0	4.5	0.3
ECO2	0	35	60	4	1
ECO3	0	25	69	5	1

## All mixtures tested in TB and **only ECO2 for aging**

# **Experimental setup**

- Two mechanical frames installed inside the GIF++ bunker  $\rightarrow$  At 3 and 6 m from the source
  - $\rightarrow$  Different requirements of collaboration members
- Gas/HV/DAQ outside the GIF++ bunker





Details of the support at 3 m from the source

View of the setups inside the GIF++ bunker

# **Experimental setup**



## Main activities since last GIF AUM



- In this talk I will give an overview of:
  - Aging results
  - Comparison of beam test data (still ongoing)

## Aging studies methodology - 1

- High voltage (HV) is switched ON to a fixed value (irradiation voltage)
- Correction for temperature and pressure variations [6] applied every minute
   → To maintain a constant effective high voltage
- The detectors are exposed to the γ flux from the <sup>137</sup>Cs source

   → Absorbed current and applied HV are logged every 30 seconds
   → Study of absorbed current stability over time
- During the weekly source-off we perform a HV scan to:
  - $\rightarrow$  Measure absorbed current without irradiation (dark current)
  - → Extract Ohmic component of dark current (see next slides) for integrated charge density calculation
- We exploit the beam test campaigns to monitor RPC performance evolution



## Aging studies methodology - 2 EP-DT RPC 2 mm single gap



- Example of dark current scan vs effective high voltage
- One scan per week during the aging studies
- Linear fit between 0 and 5 kV to extrapolate Ohmic component of the dark current at the irradiation voltage
  - $\rightarrow$  This current does not necessarily flow through the gas

 $\rightarrow$  Subtracted from the current absorbed under irradiation to calculate the integrated charge density

- Trend of monitored current (fixed HV) as a function of time
- Blue curve = total current
- Red curve = total current minus Ohmic component of dark current
- Weekly source-off scans to subtract Ohmic current each week
- HV scans during aging studies = change in absorbed current



## Aging studies results – ECO2

• Aging with ECO2 (60%  $CO_2$  and 35% HFO) at ~1 vol/h



- Irradiation voltage set to 10.6 kV (8.8 for 1.6 mm SHiP)  $\rightarrow$  Source OFF knee to limit the absorbed current for long periods of time
- Most of the irradiation the ABS is 2.2 (~500 Hz/cm<sup>2</sup> background rate)
- Stability of absorbed current over time  $\rightarrow$  Spikes when HFO bottle close to the end  $\rightarrow$  investigate with GC this week

#### Aging studies results – ECO2 CMS RPC 2 mm gap ~3 m from source



- Resistivity values normalized to 20 °C
- TW shows lowest absorbed current and highest resistivity  $\rightarrow$  Lower current drawn
- Increasing trend of the resistivity over time, effect under investigation

## Aging studies results – ECO2

• Total and Ohmic dark currents at working point and Ohmic current @ 4 kV



- Up to ≈ 100 mC/cm2 of integrated charge (almost) all detectors present currents basically stable with time.
- After  $\approx$  100 mC/cm2 of integrated charge most detectors show the current  $\rightarrow$  Fluctuations and slow rise with time.
  - $\rightarrow$  Behaviour similar in all detectors under test
- For some detectors the effects is not visible in the Ohmic part  $\rightarrow$  under investigation

#### **Integrated charge progression - 1**

• Status of charge integration for all the RPC ECOgas@GIF++ detectors



- Different maximum values of integrated charge reached by the different RPCs
  - $\rightarrow$  2.5 years of irradaition
  - $\rightarrow$  Efficiency corresponding to irradiation voltage is not the same on all detectors + different distances from the Cs source
- Results obtained with subtraction of Ohmic dark current

#### **Integrated charge progression - 2**

• Status of charge integration for all the RPC ECOgas@GIF++ detectors



- Different maximum values of integrated charge reached by the different RPCs
  - $\rightarrow$  2.5 years of irradaition

 $\rightarrow$  Efficiency corresponding to irradiation voltage is not the same on all detectors + different distances from the Cs source

• Results obtained with subtraction of Ohmic dark current

## **RPC response evolution during aging ALICE**

• Aging test with ECO2 (35/60 HFO/CO2) gas mixture ongoing since 2022

Example for STD gas mixture (ECO2 under investigation)



- Threshold is comparable between 2022 and 2024
- Slightly larger prompt charge in 2024
   → Similar large-signal fraction
- Can be explained by larger average signal amplitude
- Slightly lower average time over threshold

 Large current drift observed only in ALICE, shift of WP

 $\rightarrow$  Effects can partly be explained by preexisting issues with the ALICE RPC (under investigation) \$16/19\$

## **RPC response evolution during aging EP-DT**

• Comparison of performance for EPDT RPC before and after the aging studies with ECO2



- Integrated charge ~115 mC/cm<sup>2</sup>
- WP increased in 2024 wrt 2023, yet (~+100 V for STD, ~+200 V for ECO2 and ~+150 V for ECO3)
- Max **source off efficiency** decreases maximum by ~2% (could be due to alignment)
- Source off large-signal **probability** reduced for all the mixtures
- Max efficiency under irradiation for same background reduced in 2024 vs 2023 for all mixtures (~2% for all mixtures)

## **RPC response evolution during aging**

EP-DT RPC source on current vs rate at WP.

Comparison between 2023 and 2024 TB

- Currents under irradiation slightly higher in 2024 wrt 2023 → Visible for all mixtures
- Increase of dark current

   → Could be related to electrode degradation
   → Chemical analyses needed
- Ratio between current and rate
  - $\rightarrow$  Estimation of total charge per gamma hit
  - $\rightarrow$  Higher in 2024 wrt 2023
  - $\rightarrow$  For all mixtures and for all ABS tested @ GIF++
  - $\rightarrow$  Partly explained by higher dark current in this detector



EP-DT RPC average charge per gamma hit for different GIF++ ABS filter. Comparison between 2023 and 2024 TB

## **RPC response evolution during aging**



- Shift of the efficiency curves (few hundreds V) towards larger HV is observed
  - $\rightarrow$  For ALL gas mixtures used (so not directly caused by the gas)
  - $\rightarrow$  Smaller for STD with respect to ECO2 and ECO3
  - $\rightarrow$  Might be caused by changes in the bakelite resistivity? (under investigation)
- Max efficiency remains approximately stable after the irradiatio

# Conclusions

- The RPC ECOgas@GIF++ collaboration is carrying out a long-term irradiation test of RPCs with different layouts using HFO/CO<sub>2</sub>-based gas mixtures
- Stability of the current absorbed with ECO2 carried out for all detectors

 $\rightarrow$  No general instability observed

 $\rightarrow$  For some RPCs (e.g. CMS), current is more stable over time. This point is under investigation

Comparison of beam test data ongoing (2022 + 2023 + 2024)

 → No appreciable decrease in maximum efficiency
 → Shift of working point observed for all detectors and all mixtures, correlation with current increase and changes in bakelite resistivity ongoing
 → We are preparing a paper to summarize all these results

# Outlook

- Plan to continue irradiation campaign throughout 2025 to accumulate more charge
- Beam tests in 2025 to further compare RPC performance throughout aging studies
- Improvements to the experimental setup to have more parameters under control to better understand the origin of the fluctuating currents observed
  - Detection of potential leakage currents
  - Provide a reading of the gas mixture humidity for each detector
  - Perform systematic ISE measurements (to monitor fluoride impurities production)

#### • Papers:

High-rate tests on Resistive Plate Chambers filled with eco-friendly gas mixtures Eur. Phys. J. C (2024) 84:300
 Preliminary results on the long term operation of RPCs with eco-friendly gas mixtures under irradiation at the CERN Gamma Irradiation Facility, accepted for publication on EPJ plus focus point on the green transition of particle detectors
 Performance of thin-RPC detectors for high rate applications with eco-friendly gas mixtures Eur. Phys. J. C (2024) 84:605
 Proceedings of the ICNFP22 conference submitted as a paper on IJMPA

#### • Prizes and acknowledgments

1) ALICE thesis award for Luca Quaglia's PhD thesis in July 2024 (for theses defended in 2023)

2) Prize for the third best contribution at the RPC 2024 conference in Santiago de Compostela

# Thank you for your attention!

# References

[1] Council of European Union, Regulation (EU) No 517/2014 of the European Parliament and of the Council of 16 April 2014 on fluorinated greenhouse gases and repealing Regulation (EC) No 842/2006 Text with EEA relevance (2014) https://eur-lex.europa.eu/legal-content/EN/TXT/?uri=CELEX%3A32014R0517

[2] A. Bianchi et al., *Characterization of tetrafluoropropene-based gas mixtures for the Resistive Plate Chambers of the ALICE muon spectrometer*, 2019, JINST 14 P11014, https://doi.org/10.1088/1748-0221/14/11/P11014

[3] B. Liberti et al., Further gas mixtures with low environment impact, 2016, JINST 11 C09012, https://doi.org/10.1088/1748-0221/11/09/C09012

[4] R. Guida et al., Performance studies of RPC detectors with new environmentally friendly gas mixtures in presence of LHC-like radiation background, Nuclear Inst. and Methods in Physics Research, A 958 (2020) 162073, https://doi.org/10.1016/j.nima.2019.04.027

[5] R . Albanese et al., RPC-based Muon Identification System for the neutrino detector of the SHiP experiment, 2023, JINST 18 P02022, https://doi.org/10.1088/1748-0221/18/02/P02022

[6] Abbrescia, M., et al.: *Operation, performance and upgrade of the CMS Resistive Plate Chamber system at LHC.* Nuclear Instruments and Methods in Physics Research Section A: Accelerators, Spectrometers, Detectors and Associated Equipment 732, 195–198 (2013) https://doi.org/10.1016/j.nima.2013.05.150605

[7] Young, C. J. et al., Atmospheric Perfluorinated Acid Precursors: Chemistry, Occurrence, and Impacts, Rev. Environ. Contam. T., 208, 1–109, (2010) https://pubmed.ncbi.nlm.nih.gov/20811862/

[8] George, C. at al., *Kinetics of mass transfer of carbonyl fluoride, trifluoroacetyl fluoride, and trifluoroacetyl chloride at the air/water interface,* J. Phys. Chem., 98, 10857–10862, https://doi.org/10.1021/j100093a029, (1994), https://pubs.acs.org/doi/10.1021/j100093a029

[9] L. M. David et al., *Trifluoroacetic acid deposition from emissions of HFO-1234yf in India, China, and the Middle East* Atmos. Chem. Phys., 21, 14833–14849, 2021, https://doi.org/10.5194/acp-21-14833-2021

#### On the HFO ecology - 1 B. Mandelli https://indico.cern.ch/event/1263322/

#### But not only detector performance...



- HFO dissociation in atmosphere might leas to the creation of TFA (toxic chemical for humans)
- Deposition on land following rain fall and consequent exposure to humans
- Studies on the matter (such as those reported in [7-9]) are not yet conclusive
- Research work on this direction is ongoing and we are studying these gases since for now they are not deemed as pollutants

#### On the HFO ecology - 2 B. Mandelli https://indico.cern.ch/event/1263322/

- PFAs: Per- and polyfluoroalkalyl substances:
  - Group of synthetic substances consisting of carbon chain + fluorine
  - Widely used in the industry and can leak into water/air/soil
  - Prolonged exposure harmful for humans
  - More than 15k PFAs identified
- Possible new regulations to ban PFAs

- Not yet clear if HFO will be included + not clear if the ban will be immediate or if derogations are foreseen

#### A possible new regulation?

#### PFAS: Per- and polyfluoroalkyl substances

- PFAS are a large class of synthetic chemicals considered environmental pollutants with links to harmful health effects.
- They all contain carbon-fluorine bonds: they resist degradation when used and also in the environment.
- Concern is growing on their use as they pollute the environment: PFAS have been frequently observed to contaminate groundwater, surface water and soil.

#### **PFAS Regulation**

- On February 7, 2023, the European Chemicals Agency (ECHA) released a proposal regarding PFAS restrictions:
  - It aims to be biggest chemical ban out of health considerations.
  - The proposal sets concentration limits below which the presence of PFAS would not be restricted: but which products?
  - None of the proposed restrictions will occur immediately: but when? Possible derogations?

