Advancing RPC Detectors with Alternative Eco-Friendly Gas Mixtures and Recuperation systems

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EP-DT

Detector Technologies







- RPCs at LHC
- Alternative gases and low-GWP gas mixtures
- Gas properties studies

• Gas Recuperation system



RPCs at LHC: GHG consumptions

RPCs at LHC are operated with 65-95% of *<u>R-134a</u>*

R-134 consumption during LHC Runs

Run 2: ~ **100 000 tCO2e**/year emitted from **R-134a** consumption Mostly due to leaks at detector level \Rightarrow reparation campaign in LS2

Environmental + Economical factors

R-134a price increased of about 2.5 times w.r.t to 2015

RPCs at LHC in Run 3

ATLAS RPC using 30% CO2 gas mixture

CMS RPC using part of recovered R-134a

Total GHG consumptions

65-95% R-134a (~2000 kg/month), 0.3-1% SF6 (~50 kg/month)





RPCs at LHC: SF6 and alternatives

SF6 use in RPCs

Electron quencher, streamer suppression **0.3%** in HPL RPCs (ALICE MID, ATLAS RPC, CMS RPC) **7%** in Glass RPCs (ALICE TOF)

Fluorinated gas with the known highest GWP ~ ${\bf 22800}$

Remarkable properties

High **dielectric strength** ~ 2.5 x Air

High **electron attachment** → captures free electron to create slow heavy ions

Chemical **inertness** \rightarrow stable molecule

Long lived in atmosphere

Non-flammable

Non-toxic

High vapour pressure at NTP \rightarrow gas at room temperature and pressure

Energy industry engineered/tested some alternatives

- 3M NovecTM gases: Novec 5110 and 4710
- HFOs and chlorinated HFOs: R-1233zd, R-1244ud
- x-methane molecules: CF3I, CCl4
- Other PFCs: C4F8O



SF6 alternatives research still ongoing in electrical industry



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Goal of RPCs studies on alternative gases



Alternatives and reduction of R-134a

RPC performance studies: experimental setup

Laboratory setup

- Single gap, 2 mm electrodes + 2 mm 80x100 cm² gap HPL
- Tests of new gases
- Gas mixtures fine tuning: up to 6 components, 0.01% precision
- Low rates, cosmic muons
- Raw waveform analysis: efficiency, st. prob., cluster size, time resolution, prompt charge

GIF++ setup

- Muon beam + ¹³⁷Cs gamma source
- Gas mixtures validation:
 - Muon beam at different background rates (ABS filters)
 - Long term studies: currents stability
 - Cosmic muons measurements





Alternative gases: R-1234ze (HFO)

R-1234ze (HFO) identified as possible replacement to R-134a

- Extremely low GWP (~ 7)
- Increasingly **wider adoption** in refrigerant industry
- However, market price and availability not yet comparable to R-134a → <u>Honeywell patented</u>
- Cannot replace 1:1 R-134a → w.p. too high → CO2/He required to lower w.p.
- Long term effects still under investigation

R-1234ze performance with CO2 (+ R-134a) with cosmic muons

- **45% HFO + CO2 (ECO1)** \Rightarrow w.p. too high (~11.6 kV)
- 25% HFO + CO2 (ECO3) ⇒ low GWP, high charge content → higher currents. Currently being tested by <u>RPC ECOGAS collaboration ⇒ see TIPP talk</u>
- 22% HFO + 22% R-134a + CO2 ⇒ higher GWP, lower charge content than HFO only. Possible compromise between performance and environment



Mid-term solution to mitigate R-134a consumption

Studies on CO2 impact when added to the standard gas mixture: 30%, 40%, 50%

- Tests performed with **muon beam** and **gamma background Apr 2022**
- Increasing CO2 increases the charge content. 30% CO2 gas mixture chosen

Detector Technologies





Gas mixture fine tuning

- **SF6 increase** to suppress streamers
- R134a/CO2/i-C4H10/SF6 64/30/5/1 chosen
- Most of foremost parameters matching those of std. gas mixture
- Background currents @ w.p. ~ 15% higher

Gas mixture started to be **used** in the **ATLAS RPC**

system for LHC run 3: Nucl.Instrum.Meth.A 1049 (2023) 168088

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Alternatives to SF6

SF6 alternatives performance

Several gas tested in laboratory

C4F8O, CF3I, R-1224YD, Novec 4710, 5110

C4F8O, Novec 5110 ⇒ performance issues

CF3I \Rightarrow safety issues (mutagenic toxicity)

Novec 4710 and **Amolea 1224yd** selected for further studies with muon beam and gamma background

GIF++ tests with muon beam and gamma background

Novec 4710 **0.1%**, Amolea 1224yd **0.5%** matching performance of Std mixture with **0.3% SF6**

Novec 4710 selected for its performance but may react with water → chemical studies ongoing
Amolea 1224yd most of them contain Cl → understanding possible pollutants formation



Aging studies

Aging studies: CO2 gas mixtures

Long term performance studies required to validate a gas mixture

Tests conducted at GIF++ with selected **R-134a/CO2 gas mixture** (HFO tests ⇒ see RPC ECOGAS talk)

Detectors constantly irradiated with **speed factor** ~ **5-10**

HV scans to **monitor detector currents** + Muon test beam when available

Target integrated charge ~ **30 mC/cm2** for **Run 3**, ~300 mC/cm2 for HL-LHC

Preliminary results

No significant variation in **Ohmic currents** @ 6 kV

Slight increase in currents @ w.p. ⇒ under investigation

No variation in muon efficiency and other foremost parameter



Gas properties studies

Chemical and physical properties of existing and new gases

Not only detector performance

Understanding physical and **chemical properties** of gases is **fundamental** to **detector** operation

Environmental chemistry helps understanding **pollutants** formation

Gas molecules interaction in the atmosphere and in the detector

Rain out
mixtures→ Water solubility → critical for humidified gas
mixturesOxidation
Photolysis
or OV (wavelength < 300 nm) → quenching
properties

UV absorption tests

Several gas tested: Novecs, HFO, N2, CO2, i-C4H10 in 190-1100 nm range

Wavelength absorption spectra in the range **100-190 nm** required for RPCs

-No commercially available instruments (custom price >= 500 kCHF)

Dedicated setup to characterize amide production in Novec 4710

Solid amide products at ambient temperature. Concentration expected to be ppb



Thanks to B. Teissandier for helping in the measurements and providing the instrument

Novec 4710 Chromatogram



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Impurities studies: F- measurements

2019 ⇒ Std., HFO + R-134a + 40% CO2

- 1. F- production does **not** depend **only** on the **currents** but also on the gap **electric field**
- Tested **R-1234ze** gas mixture produced **4x more F-** than std. gas mixture

2022 ⇒ Std, Std + CO2 (+1% SF6), HFO + R134a + 50% CO2

- 30% of CO2 in the standard gas mixture has the same F- production → F- production not proportional to R-134a
- Using 1% SF6 to 30% CO2 + R-134a increases the
 F- production → under investigation
- 5. HFO + R134a + 50% CO2 produces **4x more Fthan Std**









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Gas Recuperation

R-134a Recuperation system

RPCs LHC are operated with gas recirculation

Max. recirculation of 90% ⇒ impurities control The remaining 10% needs to be fresh gas injected ⇒ gas is exhausted to atmosphere

R134a recuperation system

Goal: <u>recover R-134a at the exhaust of RPC gas</u> <u>system</u>

First prototype developed in 2019

2020-2022 R&D on first prototype to address azeotropic gas mixture:

R-134a forms an azeotrope with i-C4H10 ⇒ **simple distillation** by boiling temperature difference **not possible**

Maximum **efficiency limited** to ~ 80%

2023: new prototype built for the CMS RPC gas system ⇒ commissioning and operation



R-134a Recuperation system

Working design

- 1.Gas mixture completely cooled down to liquid in a "cold" buffer
 2.Liquid mixture slowly heated up to gas into a "warm" buffer
- 3.Small thermodynamic equilibrium steps between vapour and liquid
- 4.**Azeotropic** vapour **exhausted** from cold buffer through a pressure controller
- 5.R-134a liquid extracted from cold buffer with a compressor
 6.Compressor stores liquid in a tank ⇒ reused from CMS RPC mixer



R-134a Recuperation system

Performance and characteristics

Number of columns = 4 Filling rate = 200-400 ln/h Extraction rate = 600 ln/h

Gas Quality monitoring from IR and GC analyses

I-C4H10 <= 0.1 % N2 ~ 150 ppm O2 ~ 30 ppm Efficiency ~70-80%

Operation and future developments

Recuperated R-134a started to be used in CMS RPC mixer Gradual increase of recuperated R-134a, from 50 to 400 ln/h Better buffer's temperature regulation for quality and efficiency

Future R&D: SF6 recuperation



Gas Analysis from warm buffer @ 400 ln/h



Conclusions

Several strategies to reduce GHG emissions from RPCs

- Intensive R&D on alternative gases
- R-134a main alternative seems to be R-1234ze
- Short-mid term solution: **addition of CO2 to std. gas mixture** ⇒ GHG and costs reduction
- SF6 alternatives are still undergoing lot of tests

Long term tests

- R-1234ze tests ongoing with RPC ECOGAS collaboration
- **CO2/R-134a** aging tests **ongoing** ⇒ no evidence of aging so far

Gas Properties studies

- R-1234ze produces more F- than R-134a
- UV tests to understand quenching properties of gases
- Novec 4710 tests ongoing to understand amide production

Recuperation gas system

- Designed for large LHC gas systems
- Current **prototype** working at **CMS RPC** with ~70% efficiency and good gas quality
- Fine tuning still ongoing to increase gas quality and efficiency





R-134a + R-1234ze + CO2/He gas mixtures @ GIF++

R-134a + R-1234ze: two gas mixtures at high rates (1 CO2 **50%,** 1 He 30%):

He gas mixture has lower working point than CO2 one

CO2 + R-1234ze gas mixtures have slightly higher efficiency drop (-2%)

He gas mixture has slightly lower currents than CO2 equivalent

Std.

HFO/R134a + CO2 50% HFO/R134a + HE 30%



Muon beam + gamma background



HFO flammability tests

Safety concerning HFO usage

• R-1234yf classified as mildly flammable → Focus on R-1234ze

R-1234ze + i-C4H10 + 40% RH flammability test conducted:

ISO 1056 standard flammability test (detachement + flame propagation criteria) performed by external company

Results

- Mixture with 1% i-C4H10 + R-1234ze is flammable
- Water vapour plays an important role

HFOs alone + i-C4H10 is flammable → Effects of the CO2 on the mixtures to be understood/checked



illustration of a flame detachment with flame propagation over a distance of at least 100 mm as criterion for flammability

test no.	lso-butane fraction in test gas mixture in mol%	fraction of test gas mixture of iso-butane and HF01234ze in mol%	fraction of air including 2.25 mol% water in mol%	reaction
9	6.2	15.0	85.0	+
10	6.0	20.0	80.0	(#)
11	4.2	13.0	87.0	+
12	3.1	10.0	90.0	+
13	2.2	13.0	87.0	. +
14	1.1	13.0	87.0	
15	1.0	10.0	90.0	+
16	0.0	12.0	88.0	•
17	0.0	11.0	89.0	× .
18	0.0	10.0	90.0	-
19	0.0	9.0	91.0	-

https://edms.cern.ch/document/2463340/1

SF6 adjustment in CO2 + R-134a gas mixture

Combination (30%, 40%, 50%) **CO2** x (0.3%, 0.6%, 0.9%) **SF6**

- Higher efficiency-streamer separation for 30%/40% CO2 + 0.9% SF6 or 30% CO2 + 0.6% SF6
 → selected gas mixtures
- Lower variation of streamer probability for the same gas mixtures





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Optimization of gas system technologies

RPCs@LHC are operated with gas recirculation

RPC validated for 90% max. gas recirculation

Higher recirculation fractions leads to accumulation of impurities

Optimization of gas systems

Finer granularity in gas distribution racks \Rightarrow ATLAS RPC, LS2

Control valves for pressure control \Rightarrow CMS RPC, LS2

Gas system modifications for 4 component gas mixtures ⇒ ATLAS RPC, Run 3

Recuperate remaining 10% of recirculated gas ⇒ CMS RPC, Run 3



ALICE MID, Run 2 ISE measurement



Waveforms of Std vs. HFO vs. HFO + R134a gas mixtures



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Reduction of R-134a: addition of non-fluorinated gas

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Reduction of R-134a in the standard gas mixture by addition of a 4th, non-fluorinated gas

O2: good performance but highly reactive → lower **flammability limit**, higher currents due to oxidation reactions

Ne: good performance but no availability on the market

CO2: good performance → **selected as main candidate** for GIF++ tests

N2: high streamer contamination at low concentrations

He: good performance but **problematic for PMTs in LHC** caverns

N2O: discrete performance but increased working point of ~ 300 V

Ar: slightly high streamer probability





