INFN BARI
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# Ageing effects in RPC detectors operated with eco-friendly gas mixtures

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on behalf of the RPC EcoGas@GIF++ Collaboration

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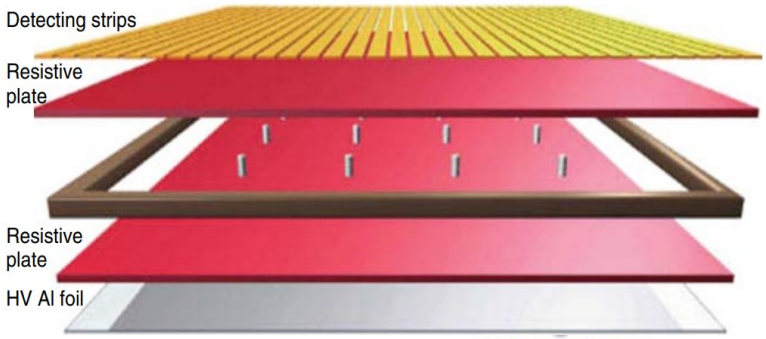
*[2] Part of this work has been funded by grant EcoRPCchem (101199027)*

# Resistive Plate Chambers




Gaseous detectors widely employed in HEP experiments for triggering and particle ID purposes:

- low cost per unit area;
- high efficiency and time resolution;
- ease of construction and robustness.


Total area currently covered by RPCs at the LHC experiments: ~$10^4$ $m^2$

RPCs successfully operated with a «standard» gas mixture, a «perfect» balance between:

***R134a (>90%) / $iC_4H_{10}$ / $SF_6$ (<1%)***

R134a and $SF_6$ are greenhouse gases with high Global Warming Potential* → $GWP_{R134a}$ = 1430, $GWP_{SF6}$ = 22800

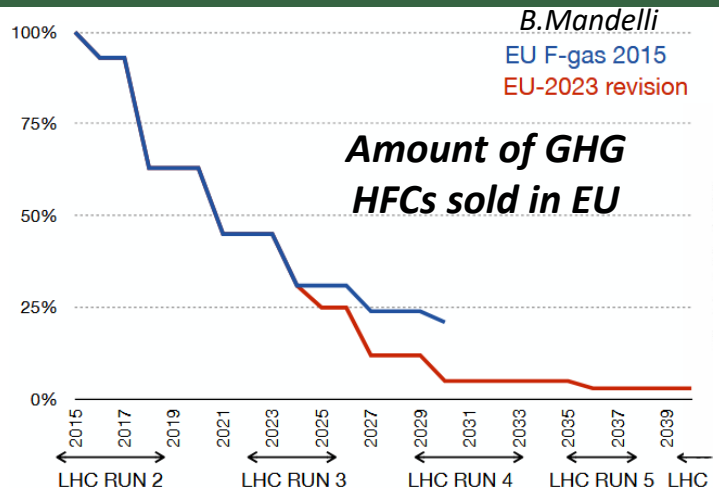
**measure of the heat trapped in the atmosphere by a ton of a given gas, if compared to a ton of $CO_2$*

# Greenhouse gases

The sale and the use of Fluorinated greenhouse gases (GHG) have been limited in EU [EU F-gas regulation 2024/573]:

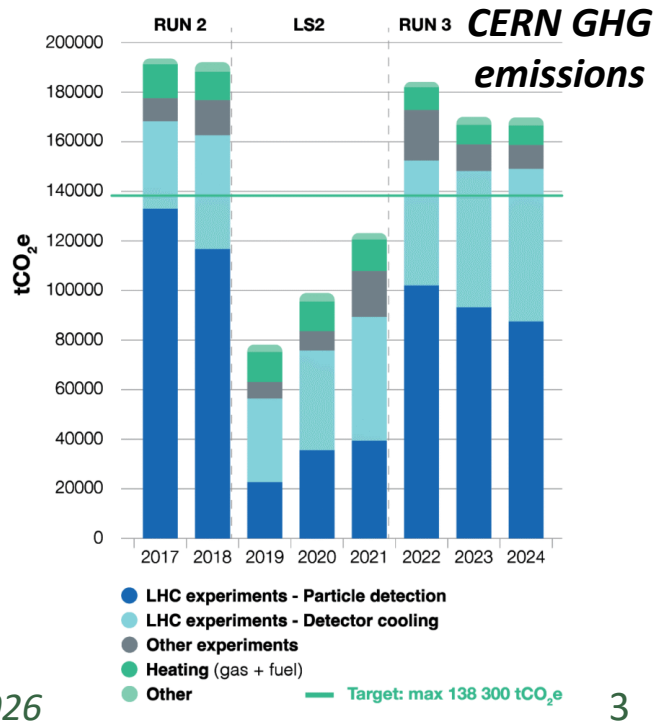
- -55% GHG emissions by 2030 w.r.t. 1990
- elimination of HFCs (such as R134a) by 2050

The Fluorinated GHG availability is being reduced and their price is increasing.




- CERN committed to reducing GHG emissions [CERN enviromental report]
  Goal: -28% by the end of LHC RUN3.
  Strategies include the **search for new eco-gas mix.**

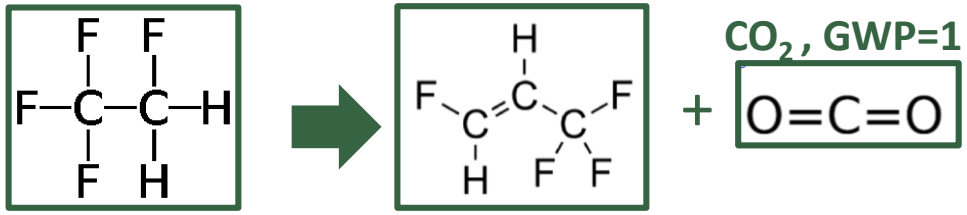
RPC groups from ALICE, ATLAS, CERN Gas team, CMS, LHCb/SHiP have joint efforts in the **RPC ECOGAS@GIF++ Collaboration** to study alternative eco-gases for RPCs with *low GWP, low toxicity and flammability guaranteeing detector performance comparable with std*



# Eco-friendly gas mixtures for RPCs

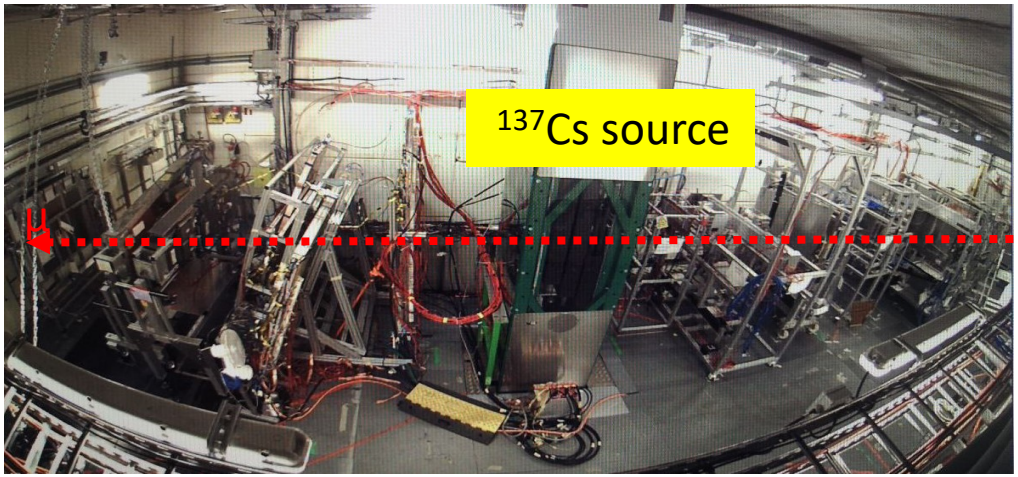
Several alternative eco-friendly gas mixtures tested. Very promising results obtained with:

**R134a, GWP=1430** **HFO-1234ze, GWP=6**




**$CO_2$, GWP=1**

***ECO2:***
***35% HFO / 60% $CO_2$ / 4% $iC_4H_{10}$ / 1% $SF_6$***
***ECO3:***
***25% HFO / 69% $CO_2$ / 5% $iC_4H_{10}$ / 1% $SF_6$***
*both with GWP reduced by 1/3 w.r.t. std*



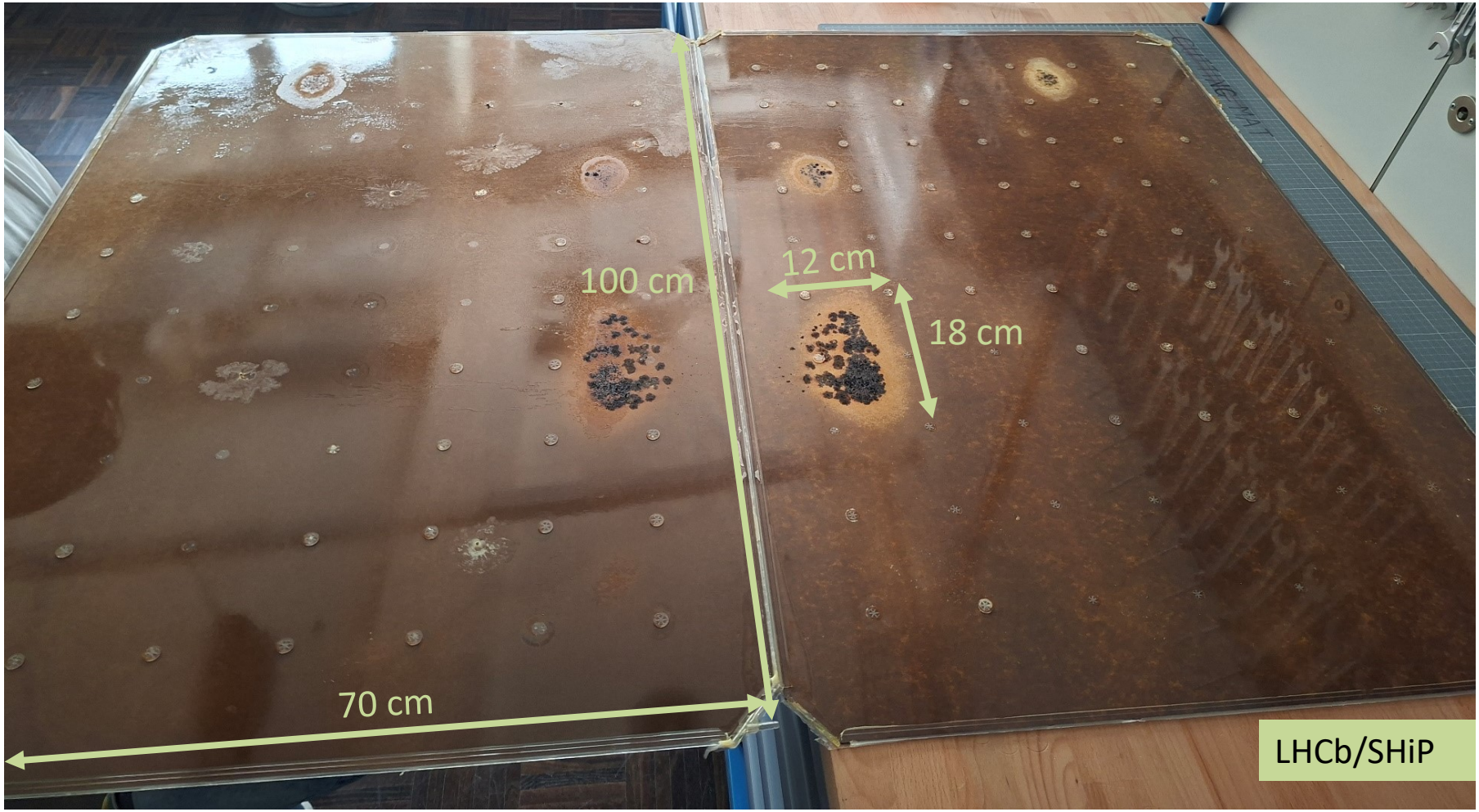

Aeging campaign started in 2022 at the Gamma Irradiation Facility (GIF++) [see Giulia's talk]:

- Different RPCs flushed with ECO2 continuously irradiated with a background rate of several hundreds of Hz/cm$^2$.
- Total integrated charge between ~100mC/cm$^2$ (ALICE) and ~350mC/cm$^2$ (LHCb/SHiP). After 50-100mC/cm$^2$, currents increased and showed fluctuations -> electrode degradation?

# Electrode surface inspection

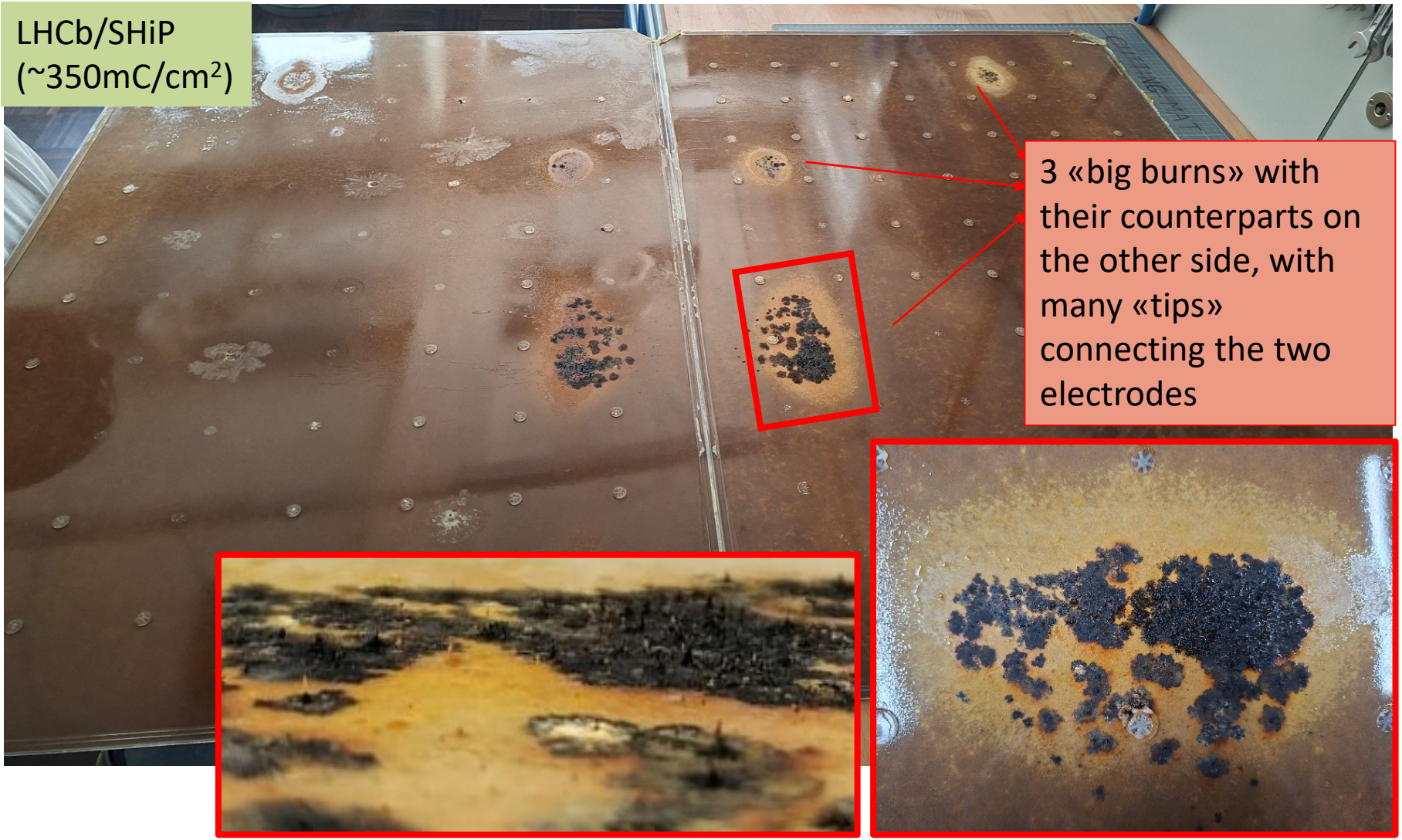
Two RPCs opened and internal HPL electrode surfaces examined.

| RPC | dimension (cmxcm) | gap thickness (mm) | electrode thickness (mm) | total integrated charge |
|---|---|---|---|---|
| ALICE | 50 x 50 | 2 | 2 | ~100 mC/cm2 |
| LHCb/SHiP | 70 x 100 | 1.6 | 1.6 | ~350 mC/cm2 |



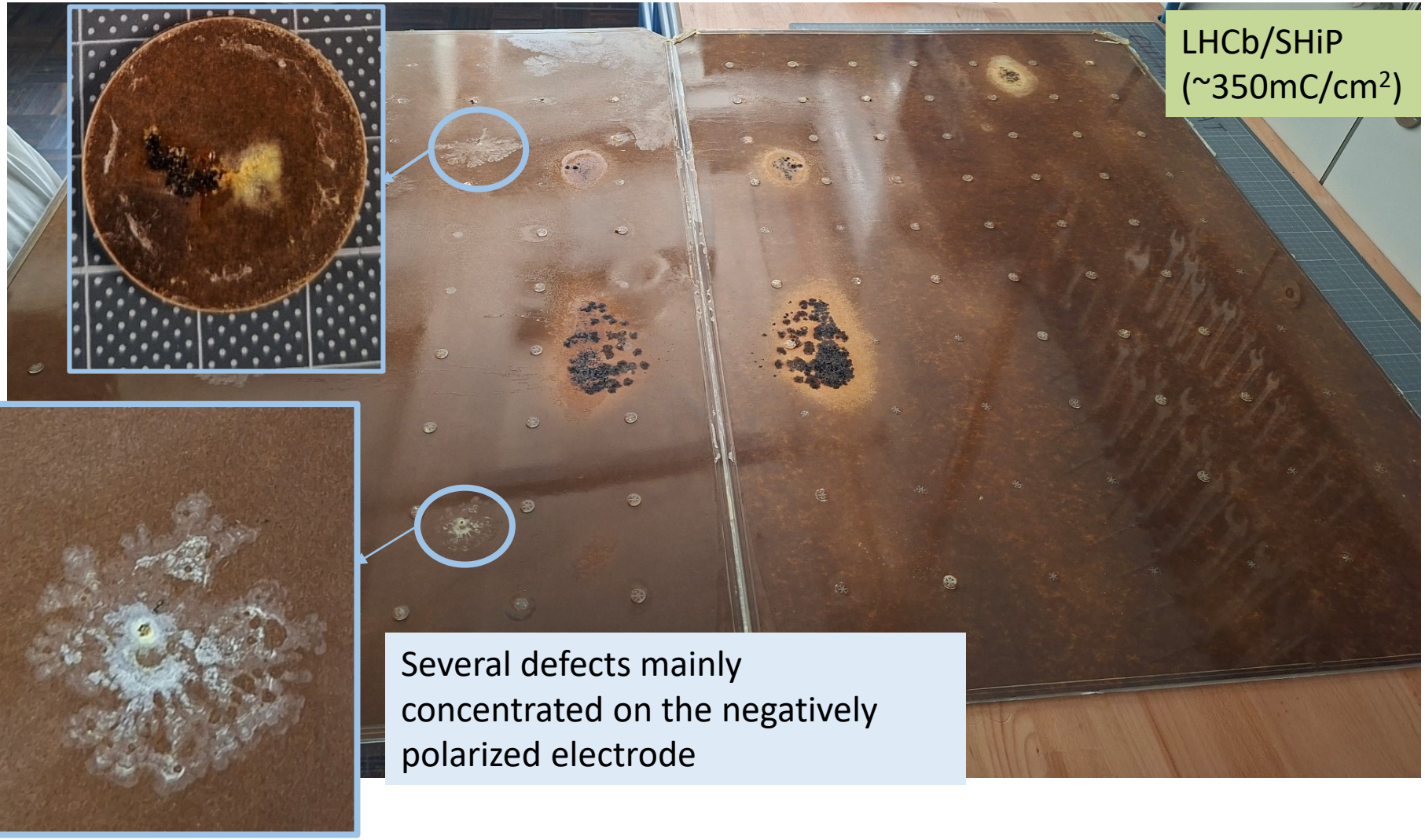
# Electrode surface inspection

LHCb/SHiP
(~350mC/cm$^2$)



3 «big burns» with their counterparts on the other side, with many «tips» connecting the two electrodes

# Electrode surface inspection



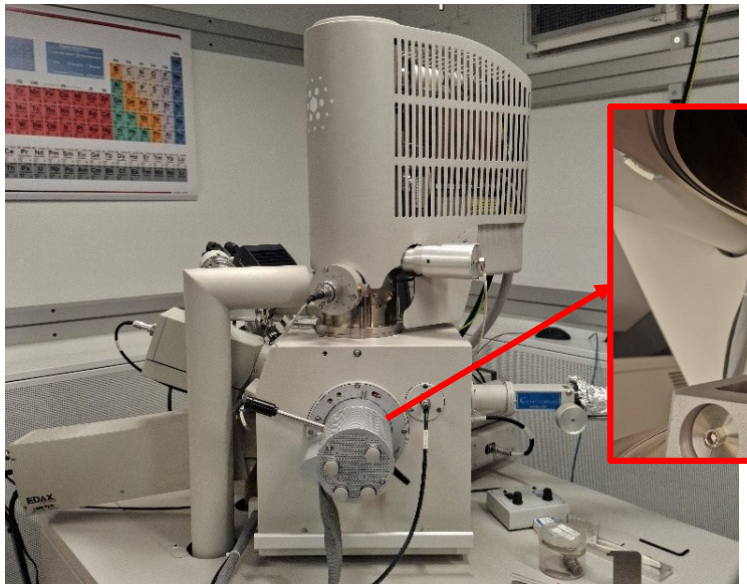
LHCb/SHiP
(~350mC/cm$^2$)

Several defects mainly concentrated on the negatively polarized electrode

Similar «defects» observed also for the ALICE RPC (~100mC/cm$^2$), without «big burns».

# SEM analysis

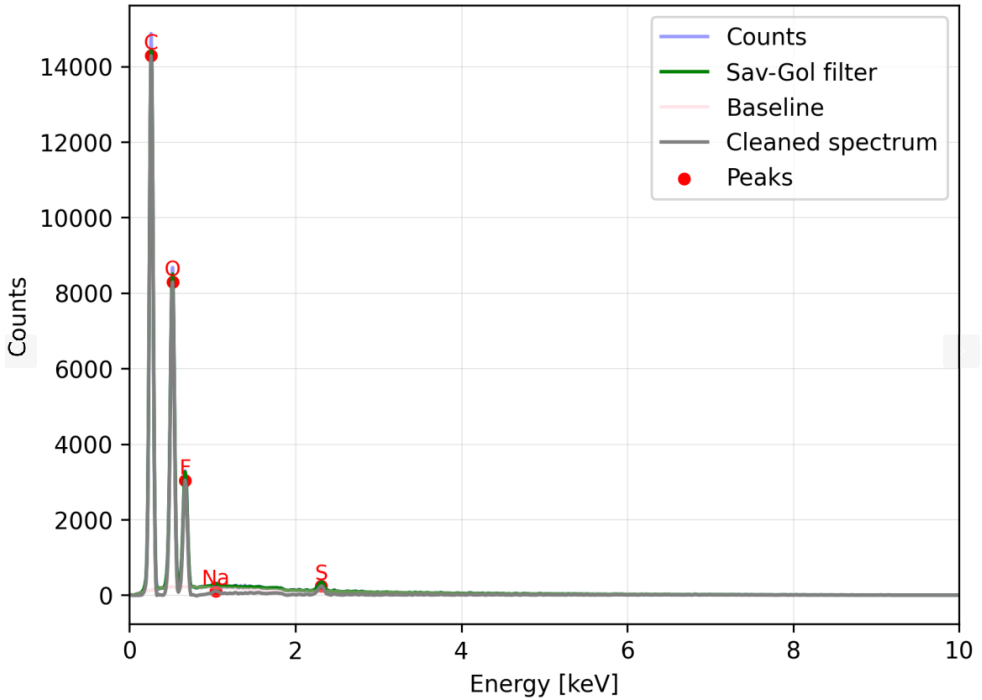
In order to analyze the morphology and chemical composition of defects and burns, analyses with Scanning Electron Microscopy (SEM) and Energy-Dispersive X-ray (EDX) spectroscopy performed.

Reference sample: at the chamber corner, not superimposed to the gap

# Spectrum analysis : methodology

- Spectrum cleaning:
  Apply a **Savitzky–Golay filter** in order to smooth noise.
  Compute and subtract the baseline using the ALS (asymmetric least squares).
- Find the peaks and local minima to define peak boundaries.
- Match peaks to elements:
  For each element, split its emission lines into K and L lines and check if detected peaks fall within ±20 eV of those lines
- Integrate peak areas and estimate relative concentrations:
  (element area / total area) * 100




What composition is expected for the electrodes?
**<u>High Pressure Laminate</u>** coated with linseed oil

*multilayer paper structure impregnated with synthetic resin:*
Main elements: **C, N, O, H**
+ traces of **S, Si, Na** due to additives used during production.

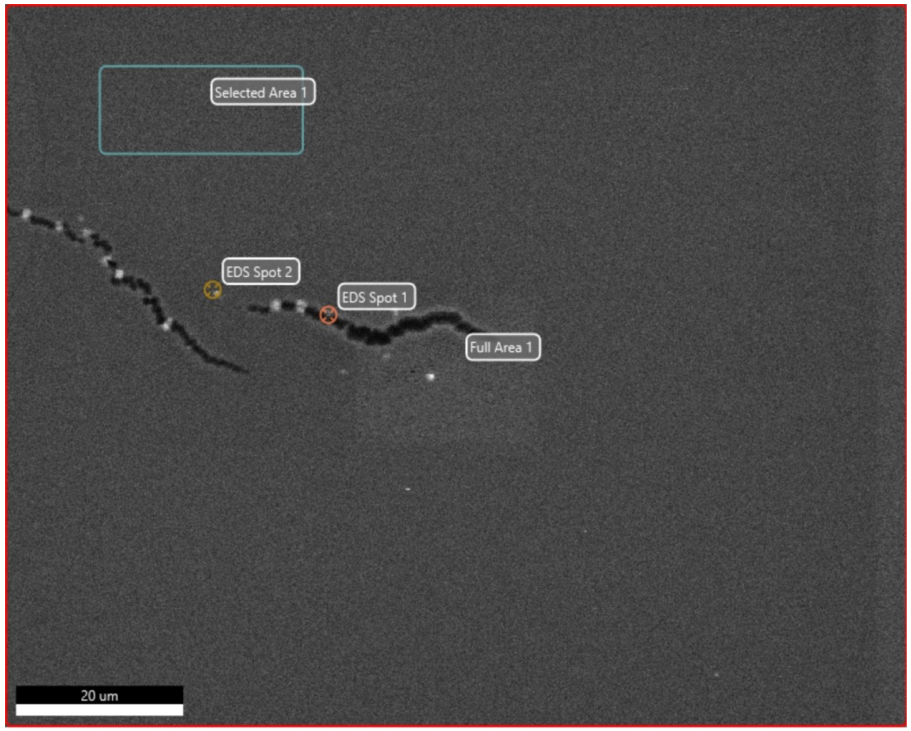
**C, O, H**

*NB H is not observable with EDX*

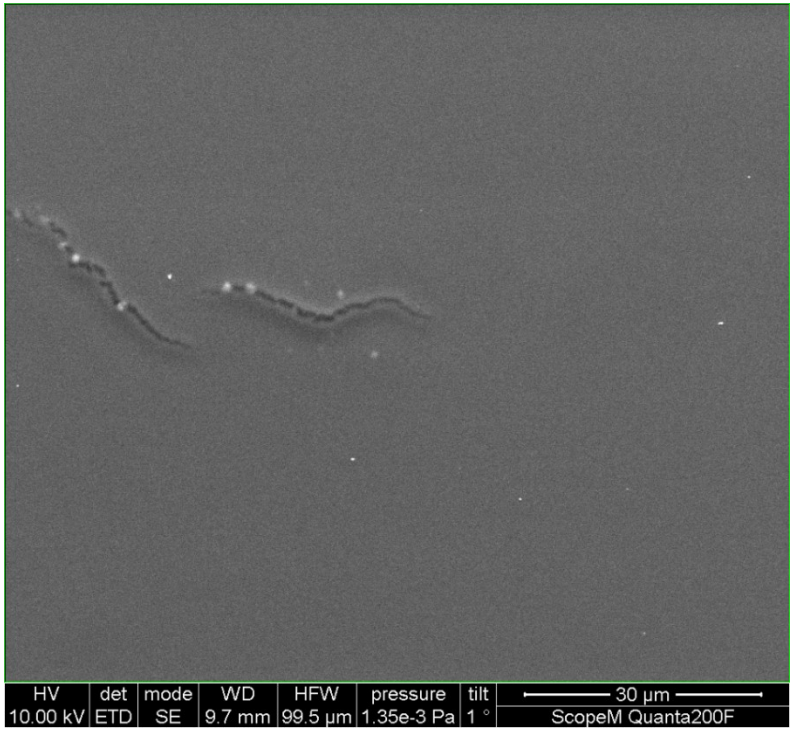
# SEM analysis - reference

Reference sample

BES mode (backscattering electrons): info on material uniformity




SE mode: info on morphology

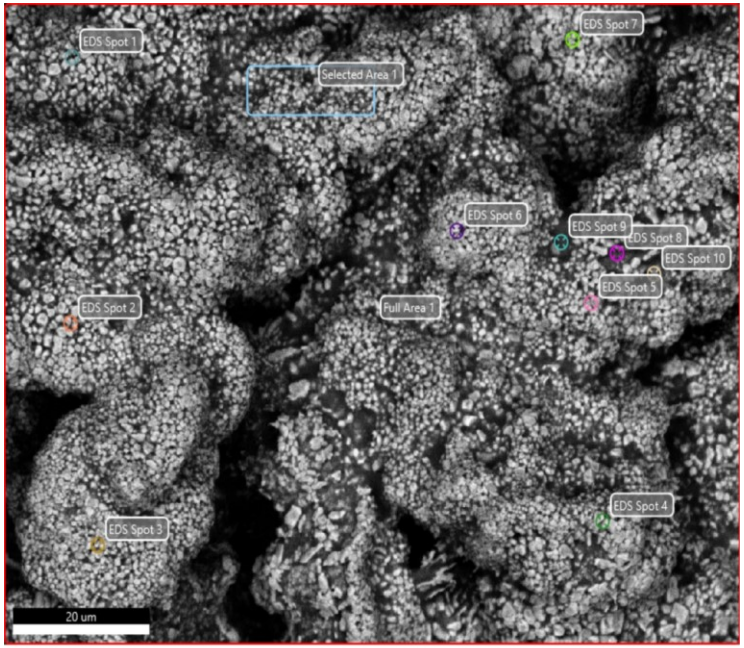


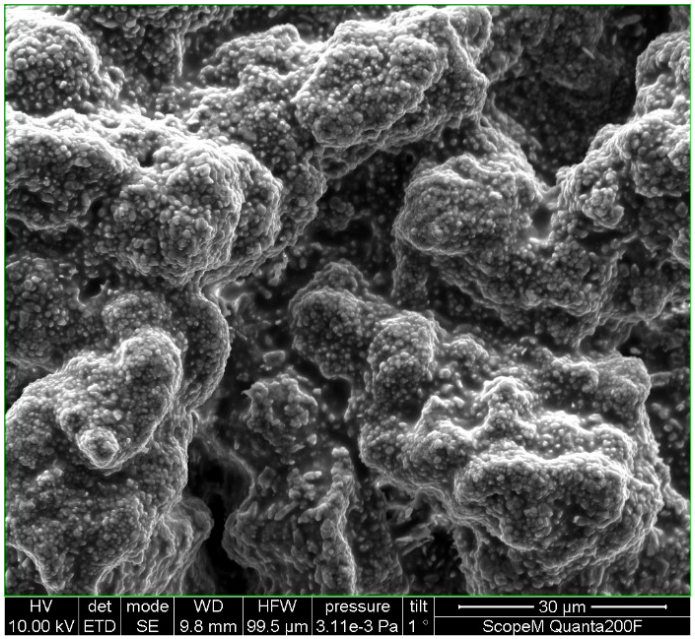

| | C (%) | N (%) | O (%) | F (%) | Na (%) |
|---|---|---|---|---|---|
| EDS spot1 | 72.1 ± 0.6 | 1.89 ± 0.07 | 18.5 ± 0.2 | 2.8 ± 0.09 | 4.8 ± 0.1 |
| EDS spot2 | 69.5 ± 0.5 | 2.70 ± 0.08 | 25.1 ±0.3 | 0.93 ± 0.05 | 1.79 ± 0.06 |

Composition as expected except for the fraction of F: gas contamination? The decomposition of F-rich gas molecules (such as HFO) could led to the production of fluoride ($F^-$).

# SEM analysis – defects (negative el.)

White powder



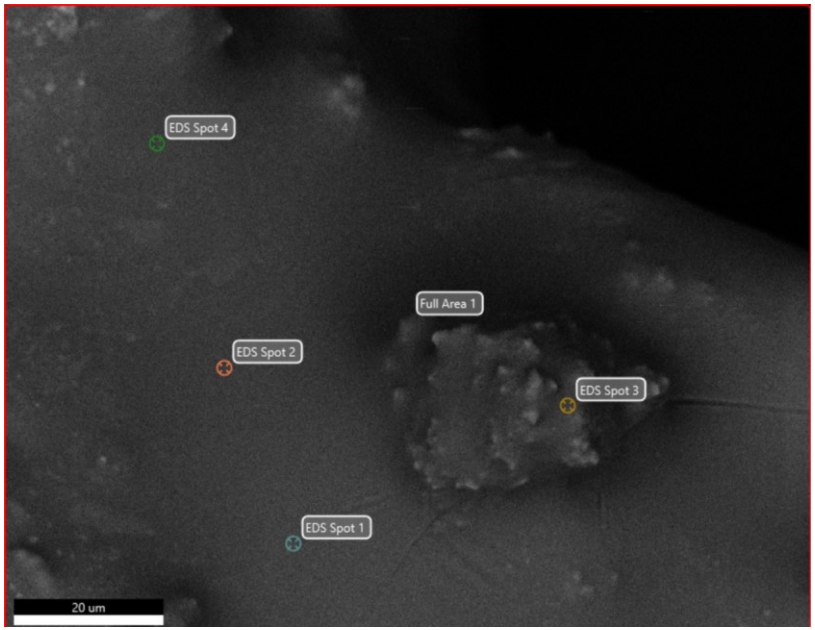

Different morphology, similar considerations for the other defects

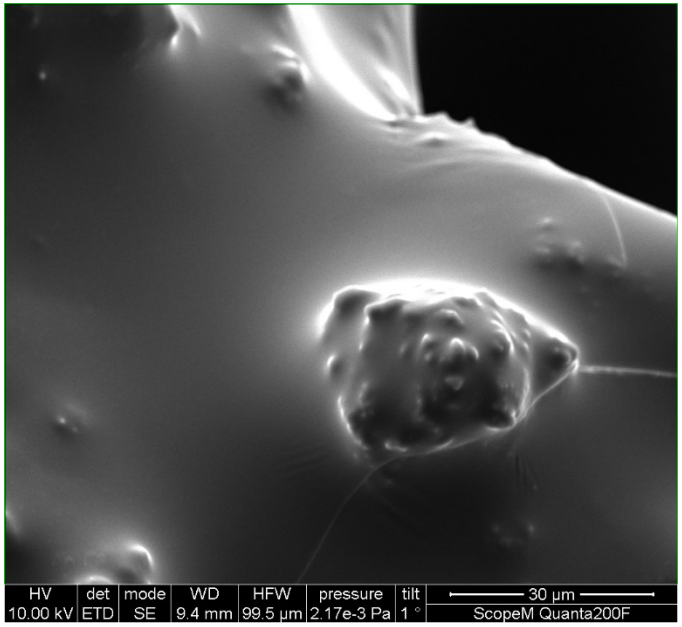
| | C (%) | O (%) | F (%) | Na (%) | Si(%) | S(%) |
|---|---|---|---|---|---|---|
| EDS spot1 | 1.10 ± 0.02 | 0.98 ± 0.02 | 62.6 ± 0.2 | 34.8 ± 0.1 | 0.279 ± 0.009 | 0.220 ± 0.008 |
| EDS spot2 | 0.72 ± 0.02 | / | 62.7 ± 0.2 | 36.6 ± 0.1 | / | / |
| EDS spot3 | 1.02 ± 0.02 | 1.04 ± 0.02 | 62.2 ± 0.2 | 35.3 ± 0.1 | / | 0.42 ± 0.01 |
| EDS spot4 | 1.53 ± 0.02 | 1.03 ± 0.01 | 53.9 ± 0.1 | 42.5 ± 0.1 | 0.62 ±0.01 | 0.43 ± 0.01 |
| EDS spot5 | 0.89 ± 0.02 | 1.14 ± 0.02 | 59.7 ± 0.1 | 37.7 ± 0.1 | 0.40 ± 0.01 | 0.139 ± 0.006 |
| EDS spot6 | 0.68 ± 0.02 | 1.07 ± 0.02 | 73.34 ± 0.3 | 24.4 ± 0.1 | / | 0.46 ± 0.02 |
| EDS spot7 | 1.55 ± 0.03 | 1.12 ± 0.02 | 55.4 ± 0.2 | 41.9 ± 0.2 | / | / |
| EDS spot8 | / | / | 67 ± 1 | 33.4 ± 0.8 | / | / |

More than 95% of the content is due to Na and F.

# SEM analysis – defects (negative el.)

burn

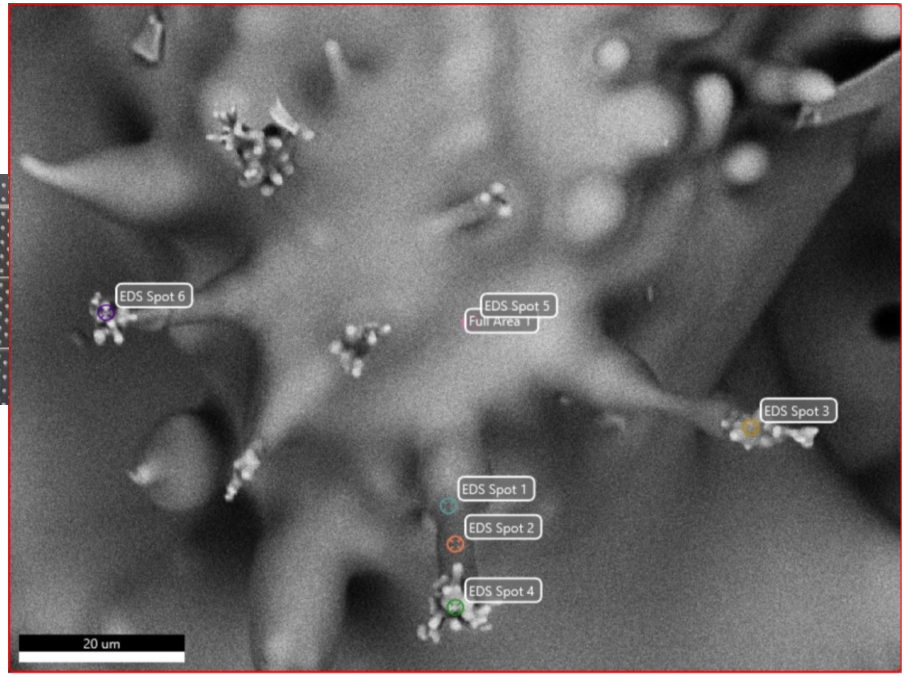


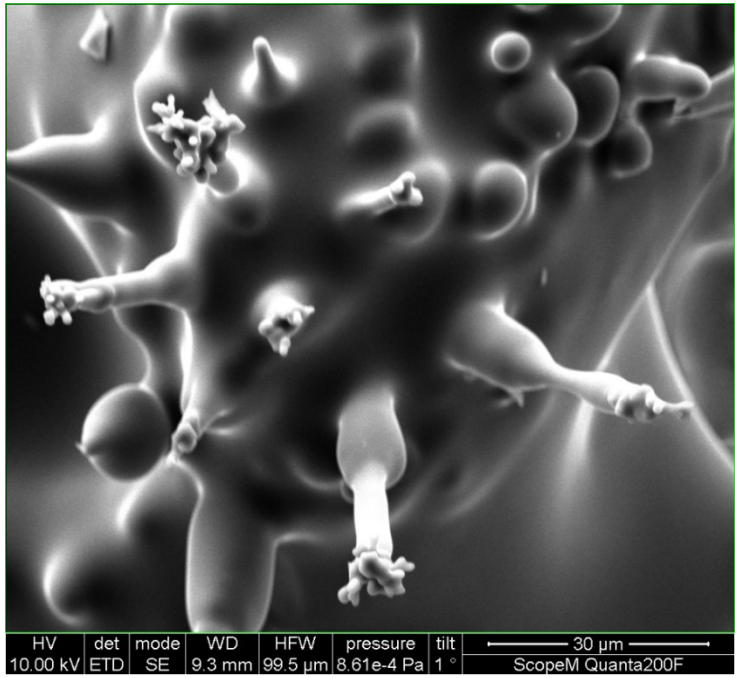




| | C (%) | O (%) | F (%) | Na (%) | S(%) |
|---|---|---|---|---|---|
| EDS spot1 | 38.4 ± 0.2 | 20.7 ± 0.1 | 36.2 ± 0.2 | 2.06 ± 0.04 | 2.71 ± 0.04 |
| EDS spot2 | 39.6 ± 0.2 | 24.1 ± 0.1 | 32.7 ± 0.1 | 1.52 ± 0.03 | 2.06 ± 0.03 |
| EDS spot3 | 37.9 ± 0.2 | 17.22 ± 0.09 | 42.2 ± 0.2 | 1.06 ± 0.02 | 1.62 ± 0.03 |
| EDS spot4 | 37.5 ± 0.2 | 21.9 ± 0.1 | 35.8 ± 0.2 | 2.68 ± 0.03 | 2.16 ± 0.03 |

Mainly C,O,F. Different morphologies observed, similar content

# SEM analysis – burns (positive el.)




## tips

| | C (%) | N (%) | O (%) | F (%) | Si(%) | S(%) |
|---|---|---|---|---|---|---|
| EDS spot3 | 71.3 ± 0.3 | 1.12 ± 0.03 | 9.00 ± 0.08 | 16.3 ± 0.1 | / | 2.28 ± 0.04 |
| EDS spot4 | 65.5 ± 0.3 | 1.25 ± 0.03 | 8.75 ± 0.09 | 21.4 ± 0.1 | 0.38 ± 0.02 | 2.79 ± 0.05 |
| EDS spot6 | 72.5 ± 0.3 | 1.19 ± 0.03 | 8.25 ± 0.07 | 16.49 ± 0.1 | 0.112 ± 0.007 | 1.44± 0.03 |

## internal

| | C (%) | N (%) | O (%) | F (%) | Si(%) | S(%) |
|---|---|---|---|---|---|---|
| EDS spot1 | 47.1 ± 0.2 | 1.89 ± 0.03 | 17.2 ± 0.1 | 32.5 ± 0.1 | / | 1.32 ± 0.03 |
| EDS spot2 | 56.3 ± 0.2 | 3.37 ± 0.04 | 15.12 ± 0.09 | 23.5 ± 0.1 | 0.189 ± 0.009 | 1.53 ± 0.03 |
| EDS spot5 | 41.8 ± 0.2 | 2.04 ± 0.03 | 23.0 ± 0.1 | 31.7 ± 0.2 | / | 1.46 ± 0.03 |

Similar composition: mainly C,O,F with C concentrated expecially on the tips.

# Conclusion and outlook

An ageing test campaign of RPCs operated with the new eco-friendly gas mixtures based on HFO/$CO_2$ was carried out from 2022 to 2025 at the GIF++:

- The total charge integrated by the RPCs ranged from ~100mC/cm$^2$ (ALICE) to ~350mC/cm$^2$ (LHCb/SHiP).
- After several tens of mC/cm$^2$, currents increased and showed fluctuations, suggesting a possible degradation of electrode surface.

ALICE and LHCb/SHiP RPCs electrodes inspected:

- Many defects observed, including “white powder”;
- Big burns found on both electrodes of the highly irradiated LHCb/SHiP RPC, including tips that could generate local short-circuits connecting the two electrodes.

Further analyses with SEM and EDX:

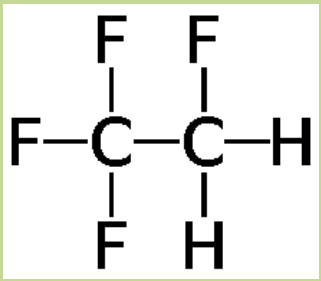
- A significant fraction of F is spotted everywhere except for ref
- White powder defects composed of Na and F (>95%)
- Burns mainly composed of C,O,F
- Different morphologies observed

Further analyses ongoing:

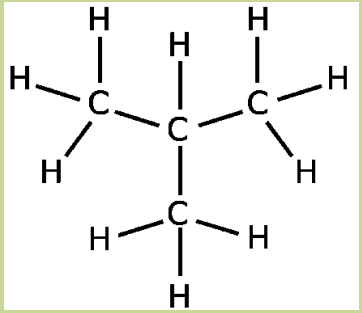
- White powder seems amorphous;
- Burns are composed of different parts: the «black» visible one is amorphous (probably C) and is embedded with further phases. XRD analysis ongoing to study their crystallographic structures.

# «Standard» gas mixture for RPCs

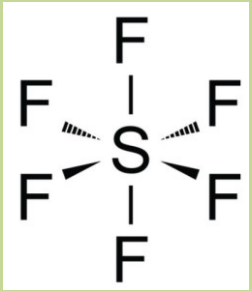

- High density of primary ion-electron pairs:
  high RPC efficiency
- Good quenching properties and electronegativity
  good rate capability and "slow" detector ageing



R134a (>90%)



$iC_4H_{10}$



$SF_6$ (<1%)

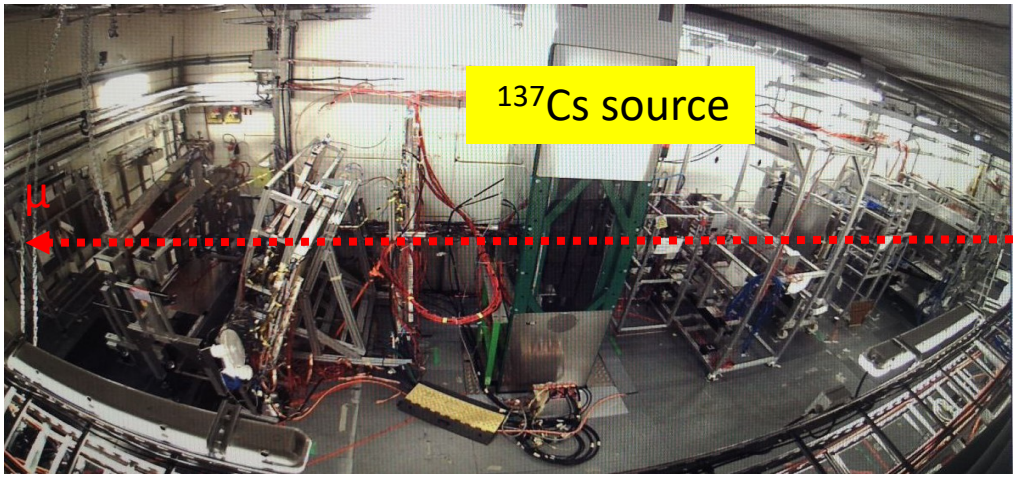
However, R134a and $SF_6$ are Fluorinated Greenhouse gases (F-gases) having high **Global Warming Potential (GWP)***
[e.g. the main component for standard gas mixture (R134a) has GWP as high as 1430]

**measure of the heat trapped in the atmosphere by a ton of a given gas, if compared to a ton of $CO_2$*

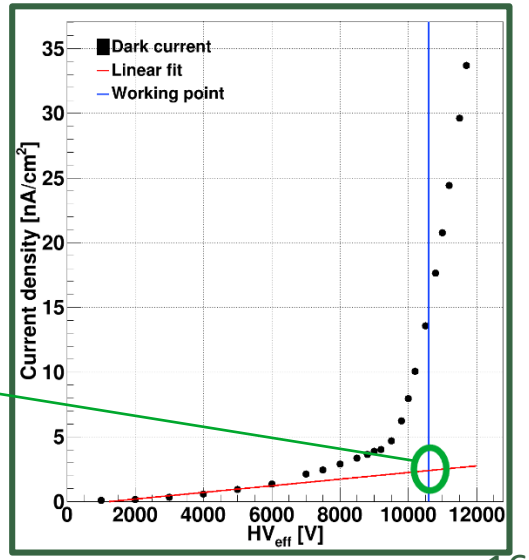
# Ageing study

In order to validate the new eco-gas, an aeging campaign was started in 2022 at the Gamma Irradiation Facility (GIF++):

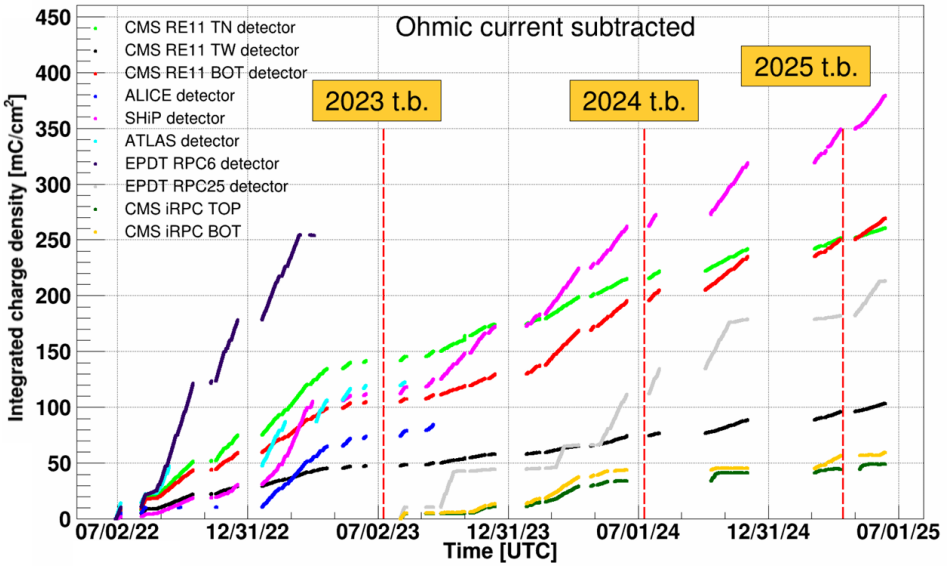
- Several RPCs with different features flushed with ECO2, switched on at fixed HV (irradiation voltage) value and continuously irradiated with a background rate of several hundreds of Hz/cm$^2$
- Weekly current vs HV scan to monitor the absorbed current without irradiation (dark current)
- Calculation of the charge integrated by each RPC under irradiation from the current flowing through the gas: $I_{tot} - I_{Ohmic}$
- Periodical beam tests with muons to assess the performance of RPCs



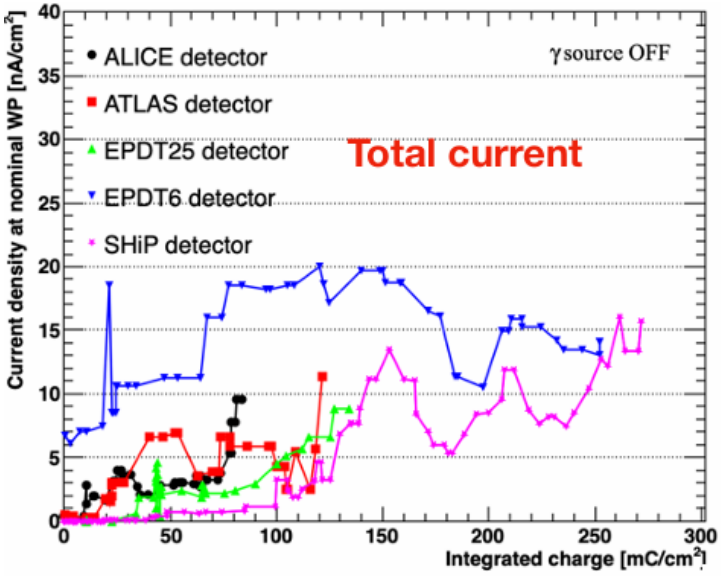

$I_{Ohmic}$ : Ohmic current (could flow through other conductive paths in the detector)

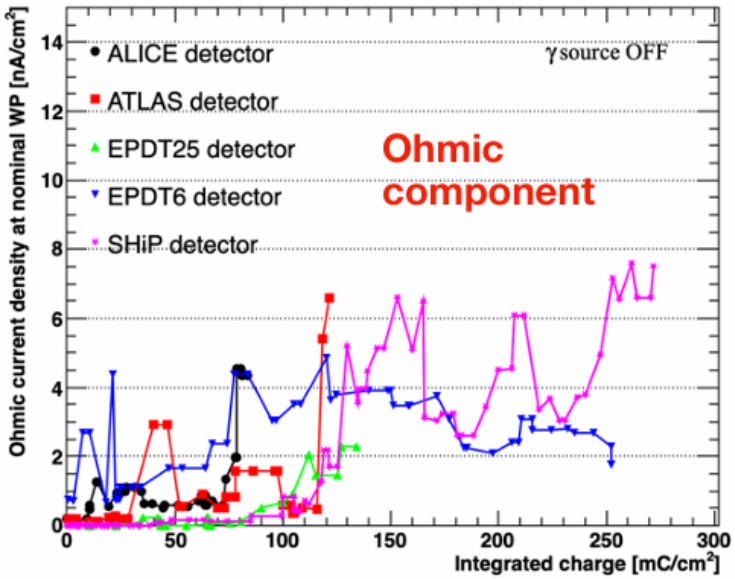


# Ageing study: results



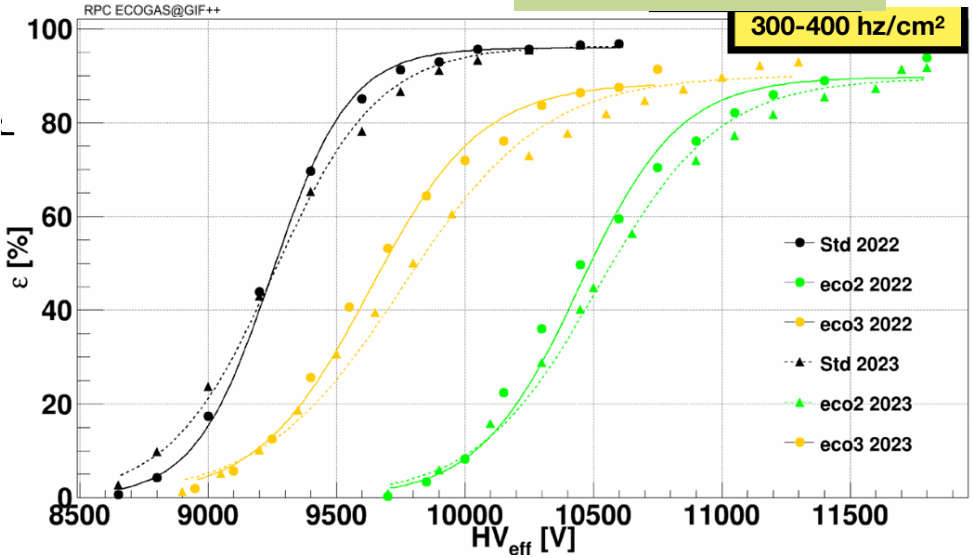

- Between ~100mC/cm$^2$ (ALICE) and ~350mC/cm$^2$ (LHCb/SHiP) integrated.
- After 50-100mC/cm$^2$ of integrated charge, both total and Ohmic currents increase and show fluctuations.
  Hint of degradation of the internal surfaces of the detectors?



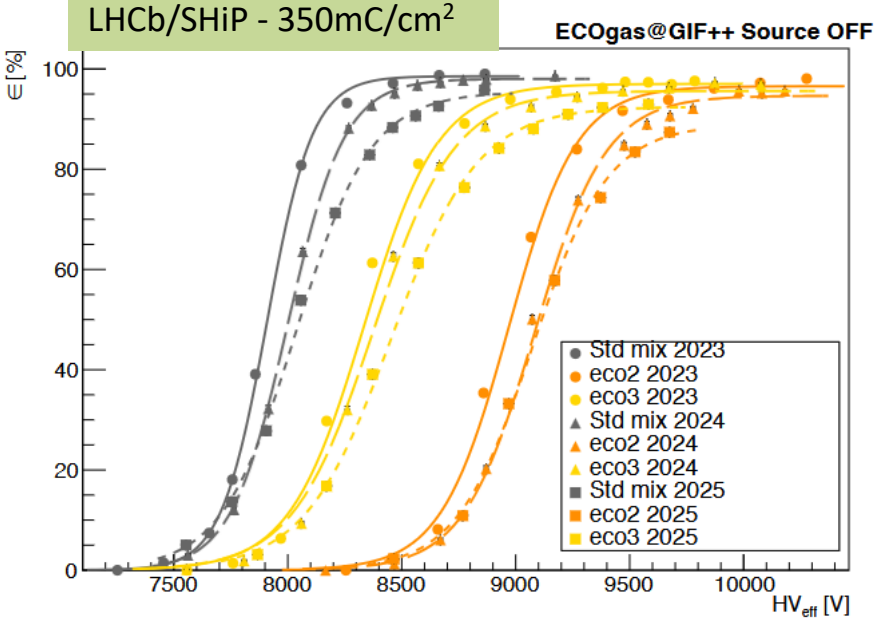



# RPC efficiency

ALICE - 100mC/cm$^2$




LHCb/SHiP - 350mC/cm$^2$



# SEM

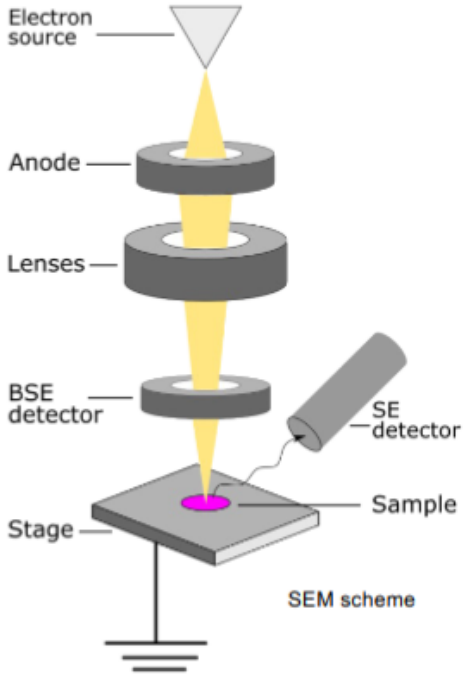
## SEM data-taking

- Electron beam to probe the material
  - Resistive materials cause charge accumulation on the sample
  - Sputter 10 nm carbon coating + connect the carbon to sample mount (grounded)

- **SE** = secondary electrons, emitted from the material (low energy ~50 eV)
  - Topography info

- **BSE** = back-scattered electrons, electrons from the beam reflected from the sample (high energy)
  - Material contrast (strong correlation with atomic number)

- **EDX** = energy-dispersive X-ray (**only for elements heavier than Li**)
  - Identify atomic composition of the material
  - Exploiting the characteristic X-ray emission of atoms

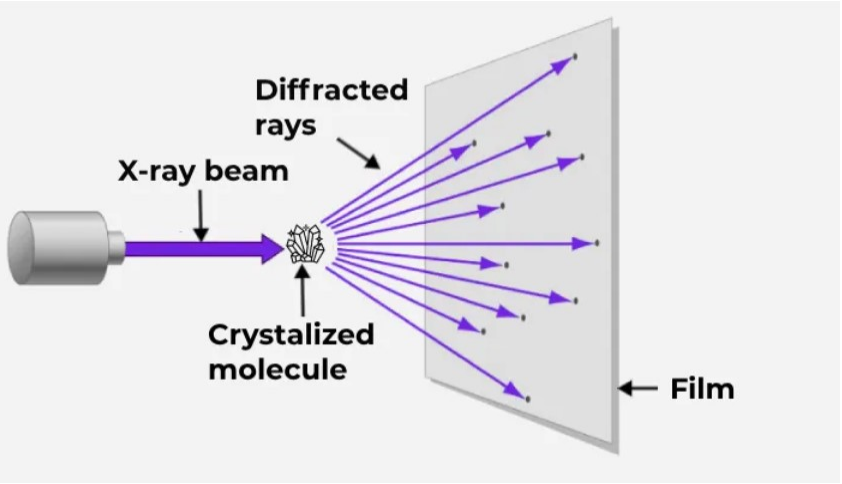


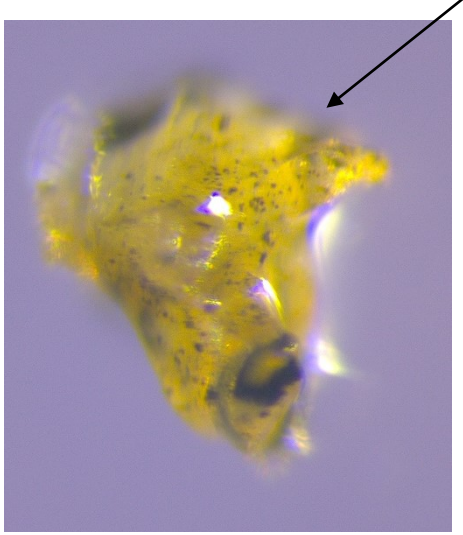

*L. Quaglia*

# XRD analysis

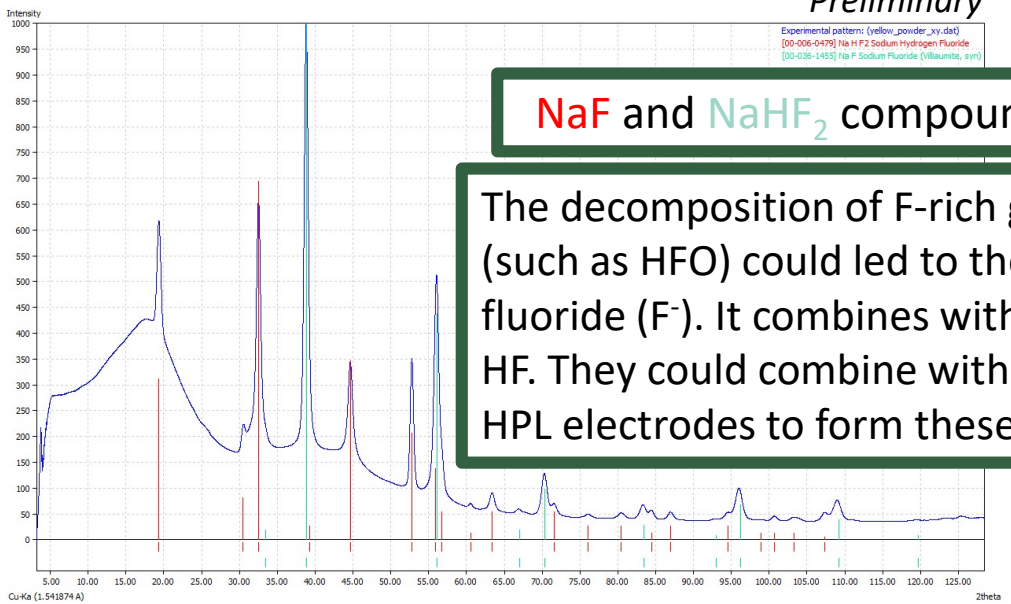
X Ray Diffraction analysis has been performed in order to analyze the different compounds:

- White powder in defects is amorphous;
- The burns are composed of different compounds. The «black» part is amorphous (probably C) and is embedded with a «yellow» part



*Preliminary*




NaF and $NaHF_2$ compounds detected

The decomposition of F-rich gas molecules (such as HFO) could led to the production of fluoride ($F^-$). It combines with $H_2O$, producing HF. They could combine with Na cointained in HPL electrodes to form these compounds.