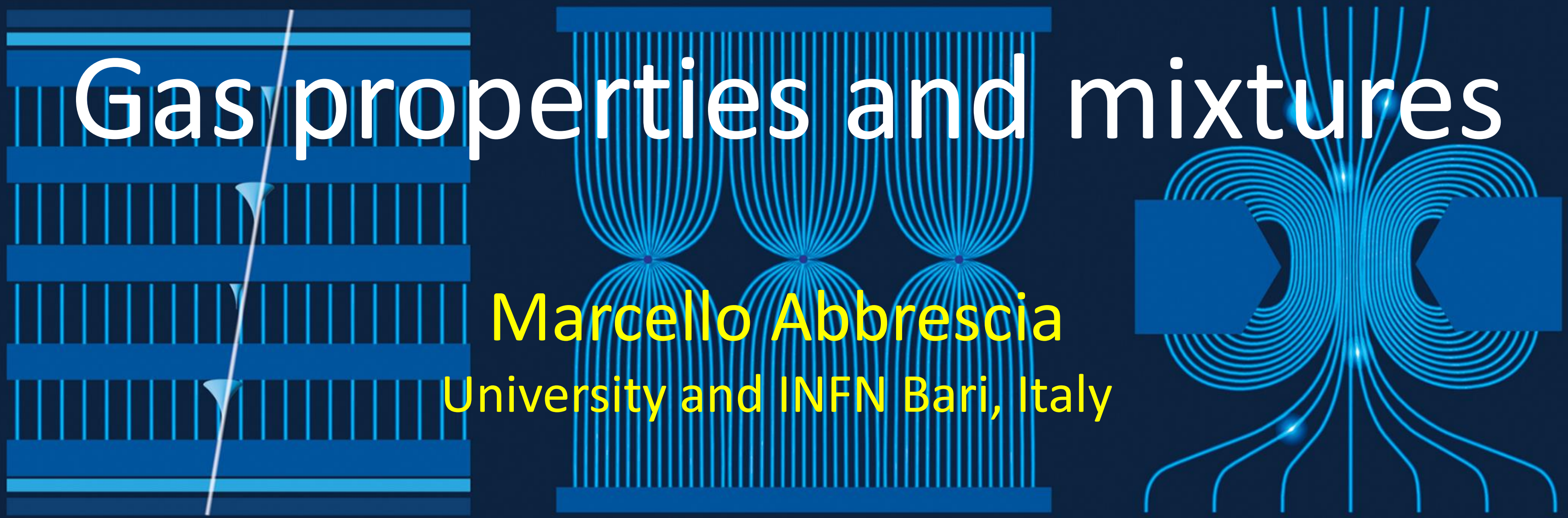


Gas properties and mixtures

Marcello Abbrescia

University and INFN Bari, Italy

DRD1 gaseous detector school
CERN, November 27 – December 6, 2024

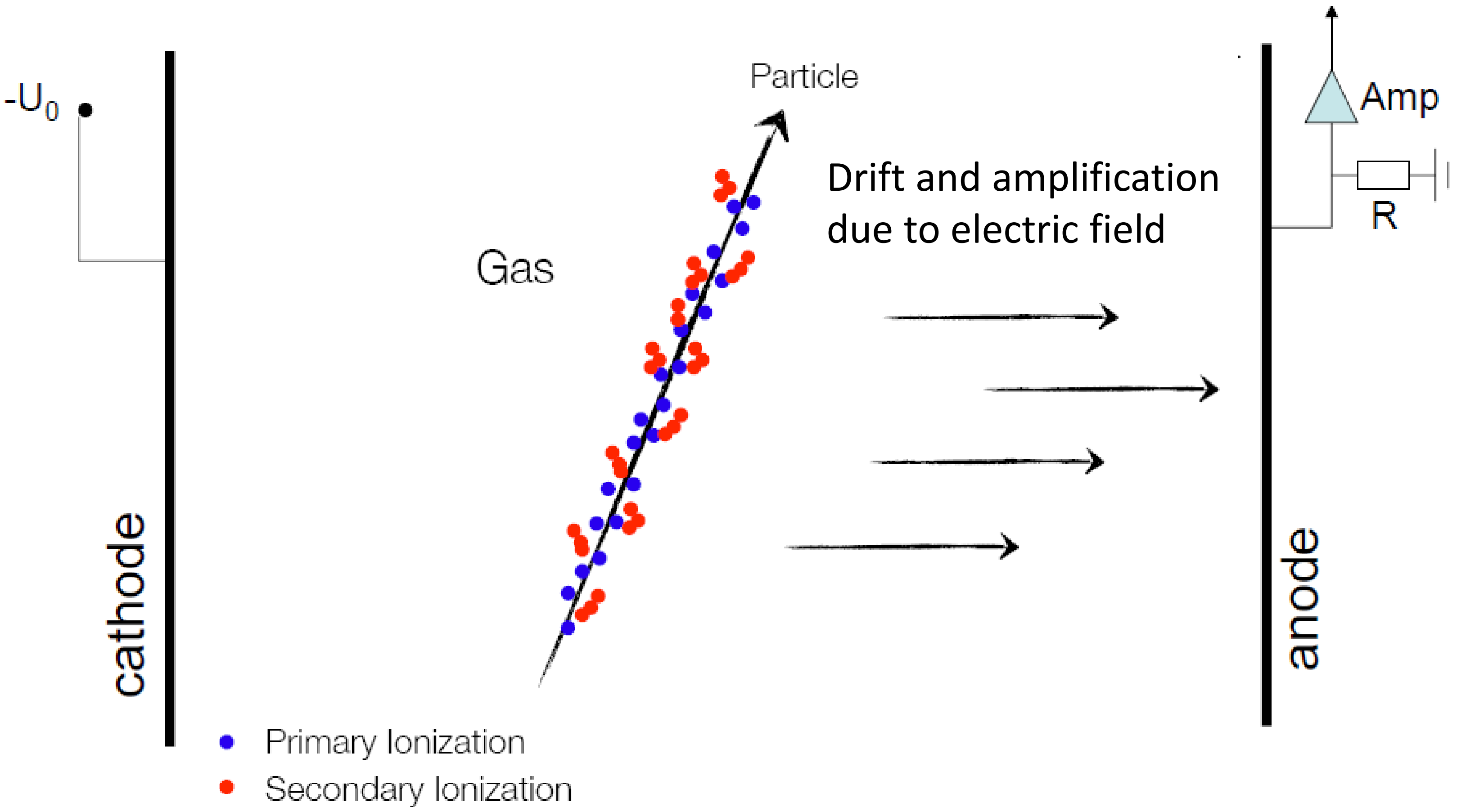
Basics

The gas (or gas mixture) is the «core» of any gaseous detector

- It is where (almost) everything happens
- Its properties contribute to determine the shape, amplitude and duration of the signals generated
  → The performance of the detector
- Aging processes in gaseous detectors strongly depend on the nature of the gas mixture used

How do we choose the «best» gas mixture?

Working principle of gaseous detectors



Drift and amplification due to electric field

Detection of an impinging particle takes place because this ionizes the gas, generating ion-electron pairs.

Ionization

Primary ionization

$$X + p \rightarrow X^+ + p + e^-$$

p = charge particle traversing the gas
X = gas atom
e^- = delta-electron (δ)

Secondary ionization

if E_δ is high enough ($E_\delta > E_i$)

$$X + e^- \rightarrow X^+ + e^- + e^-$$

Ionization energy : I_0
Average energy/ion pair : W_i
Average number of primary ion pairs [per cm] : n_p
Average number of ion pairs [per cm] : n_T

$$\langle n_T \rangle = \frac{L \cdot \left\langle \frac{dE}{dx} \right\rangle_i}{W_i}$$

[about 2-6 times n_p]
[L: layer thickness]

Not the whole energy lost by the impinging particle goes in ionization processes; a part (roughly half) goes in excitation of the gas molecules.

→ The average energy W_i needed to create an ion-electron pair is ≈ 2-3 I_0

Interest in maximizing ionization processes

Statistics of ion-electron pair production

Primary ionization events are independent and therefore follow Poisson statistics.

- Each time an impinging particle crosses a layer of gas, it has a certain probability to produce a certain number of primary electrons

- If n_P is the average number of primary electrons (ions), the probability $P(k)$ to have k electrons (ions) is:

$$P(k) = e^{-n_P} \frac{n_P^k}{k!}$$

Therefore there is always a finite probability to produce no electron-ion pairs:

$$P(0) = e^{-n_P} \quad \Longrightarrow \quad \varepsilon_{\text{det}} = 1 - P(0) = 1 - e^{-\bar{n}}$$

→ In the other cases the impinging particle is not revealed

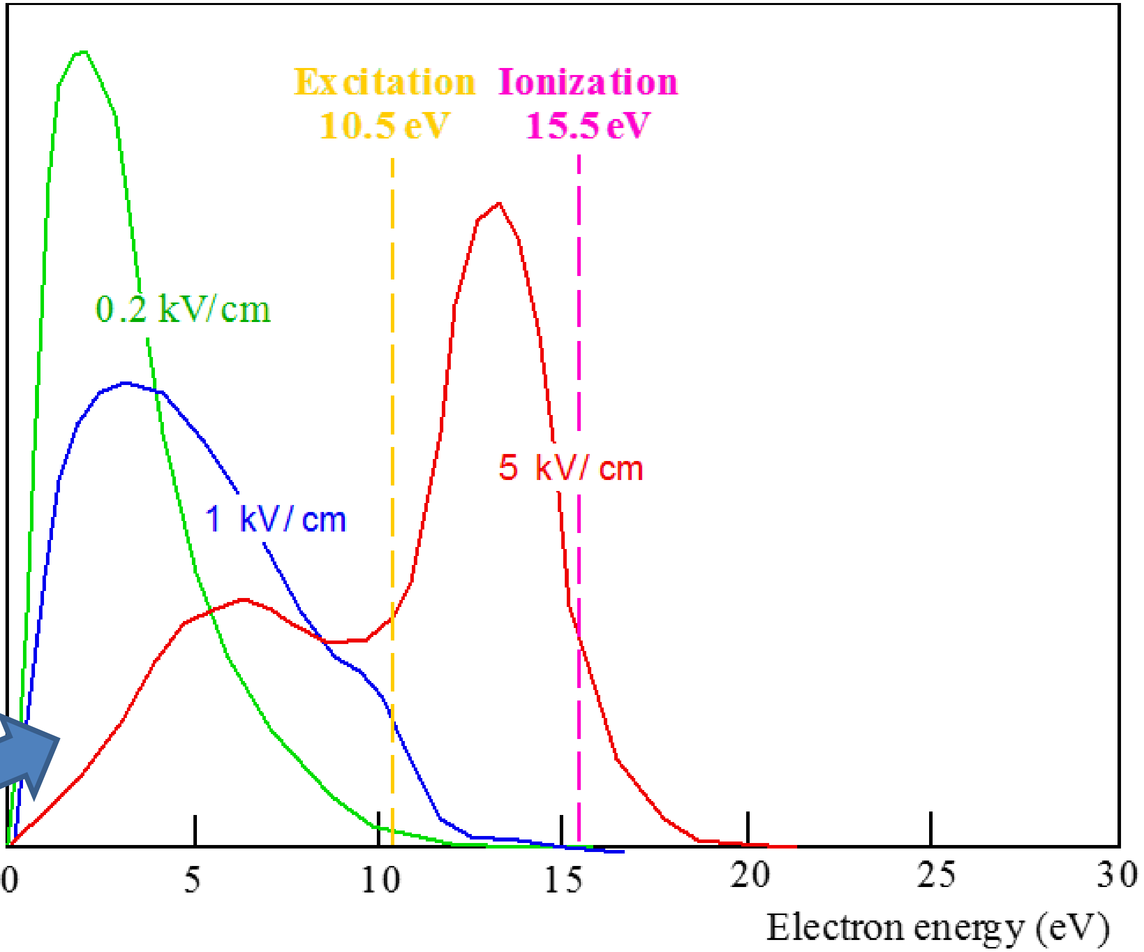
→ Intrinsic inefficiency of the detector

Note: of course n_P is proportional to the gas layer thickness and the energy loss
→ Limit on the thickness of the gas volumes that can be used.

High **E** fields: excitation and ionization

- When an applied electric field exceeds few kV/cm, more and more electrons can reach enough energy to produce excitation and ionization.

- The plot shows the energy distribution in Ar for three different values of the electric field, compared to the energy needed for excitation and ionization



Multiplication processes

Multiplication processes

- ✓ In gaseous detectors typically few tens of ion/electron pairs are produced by a MIP in few mm of gas;
- ✓ To be compared to ≈24.000 hole/electron pairs produced in 300 μm of Si.

Multiplication processes (a la Townsend) are fundamental in gaseous detectors

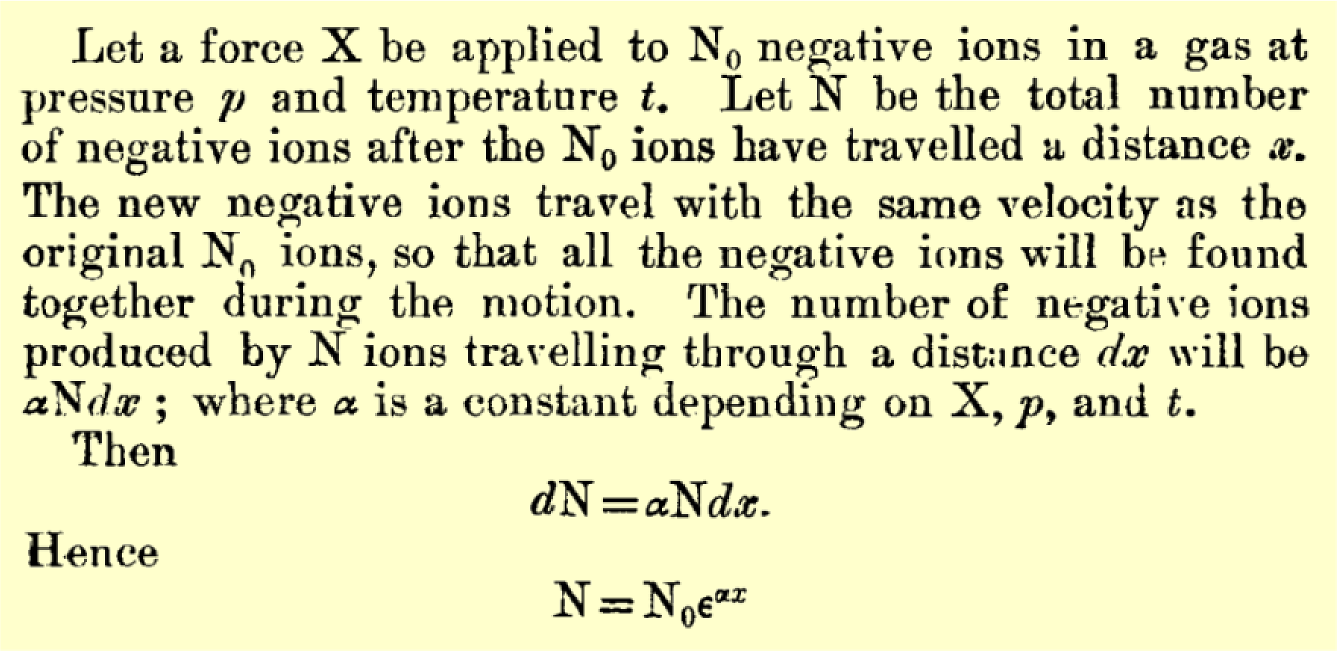
→ Because there's no hope to distinguish the signal generated by few tens of electrons from all sources of noise

All these considerations leads to the necessity of optimizing (namely enhancing as much as possible) primary and secondary ionization in gaseous detectors.

Sir John Sealy Edward Townsend
(1868-1957)

1901: Gas multiplication

- John Townsend:



Let a force X be applied to N_0 negative ions in a gas at pressure p and temperature t. Let N be the total number of negative ions after the N_0 ions have travelled a distance x. The new negative ions travel with the same velocity as the original N_0 ions, so that all the negative ions will be found together during the motion. The number of negative ions produced by N ions travelling through a distance dx will be $\alpha N dx$; where α is a constant depending on X, p, and t.

Then

$$dN = \alpha N dx.$$

Hence

$$N = N_0 \epsilon^{\alpha x}$$

[J.S. Townsend, “*The conductivity produced in gases by the motion of negatively charged ions*”, Phil. Mag. **6-1** (1901) 198-227. If access to the Philosophical Magazine is restricted, then consult a German-language abstract at http://jfm.sub.uni-goettingen.de/.]

Choice of the gas filling

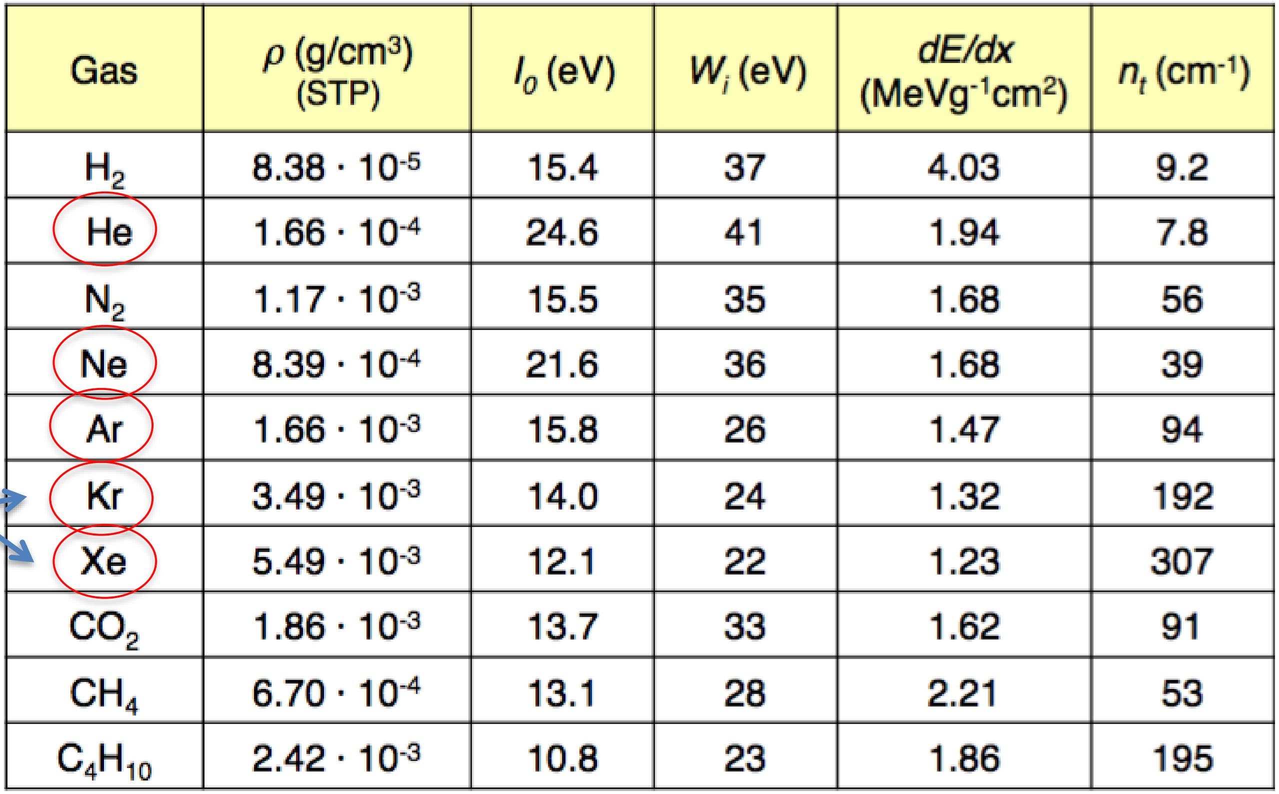
1. **Noble gases** are preferentially used due to the absence of vibrational and rotational states → **ionization dominates.**

(Polyatomic gas have non-ionizing de-excitation modes available)

→ Possible electron multiplication at low electric field

→ Choose cheap noble gases with low ionization potential

Expensive and rare



Gas	ρ (g/cm^3) (STP)	I_0 (eV)	W_i (eV)	dE/dx (MeVg^{-1}cm^2)	n_t (cm^{-1})
H_2	$8.38 \cdot 10^{-5}$	15.4	37	4.03	9.2
He	$1.66 \cdot 10^{-4}$	24.6	41	1.94	7.8
N_2	$1.17 \cdot 10^{-3}$	15.5	35	1.68	56
Ne	$8.39 \cdot 10^{-4}$	21.6	36	1.68	39
Ar	$1.66 \cdot 10^{-3}$	15.8	26	1.47	94
Kr	$3.49 \cdot 10^{-3}$	14.0	24	1.32	192
Xe	$5.49 \cdot 10^{-3}$	12.1	22	1.23	307
CO_2	$1.86 \cdot 10^{-3}$	13.7	33	1.62	91
CH_4	$6.70 \cdot 10^{-4}$	13.1	28	2.21	53
C_4H_{10}	$2.42 \cdot 10^{-3}$	10.8	23	1.86	195

Choice of the gas filling

1. Noble gases are characterized by the largest I_0/W_i ratio
2. In principle noble gases with large Z should be preferred, because of their large dimensions, leading to larger n_t/cm

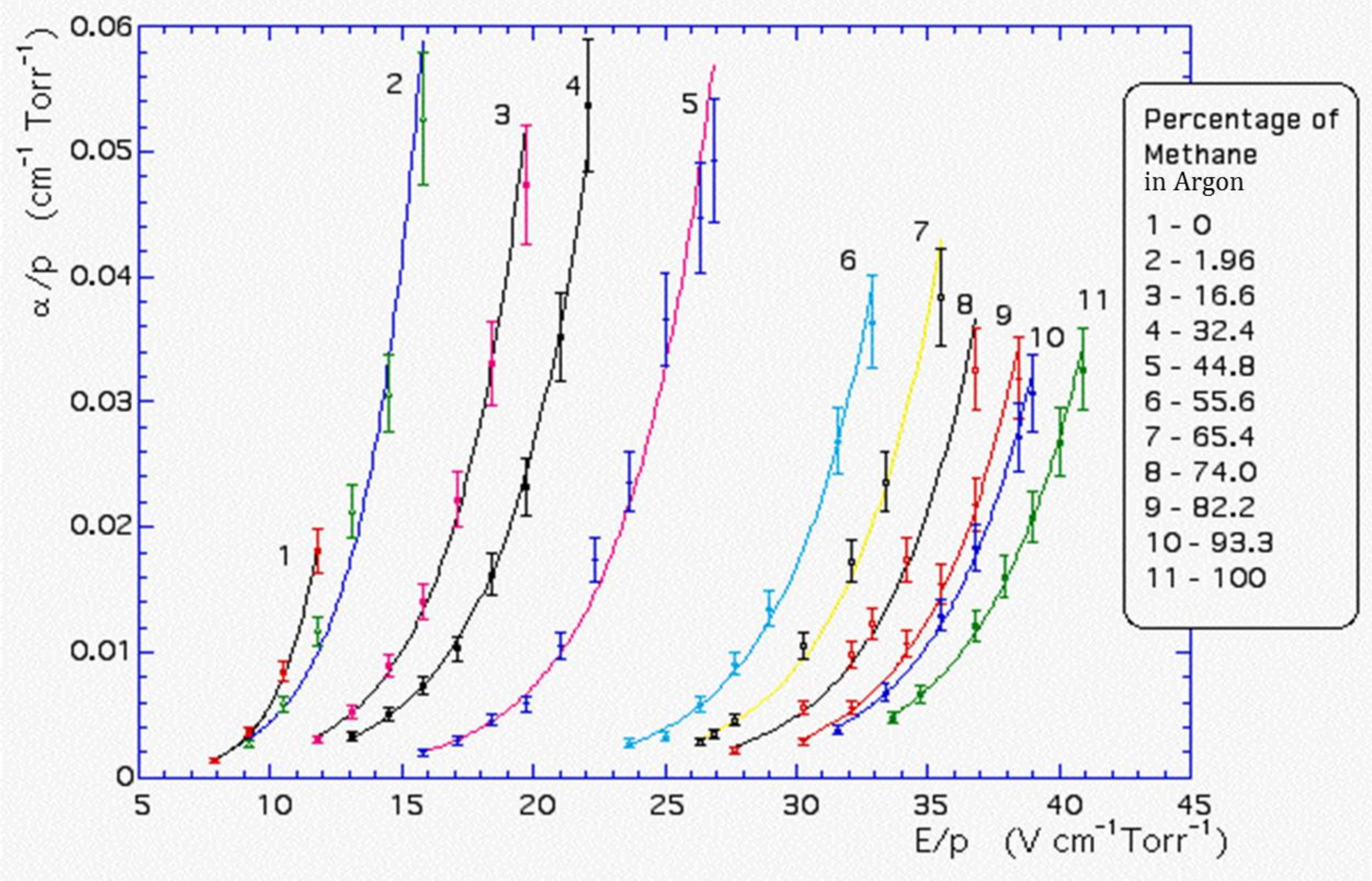
→ Kr and Xe would be ideal choices, but their drawback is their cost

→ Rn is radio active

Hence Ar was (and still is) a very popular choice

In principle one could release the requirement on I_0/W_i in the case of large molecules, characterized by large n_t/cm
Large hydrocarbons would be suitable
→ But they are flammable

First Townsend coefficient for Ar/CH_4 mixtures



The first Townsend coefficient represents the average number of pairs produced per unit length by an electron drifting in the gas

Not only ionization…

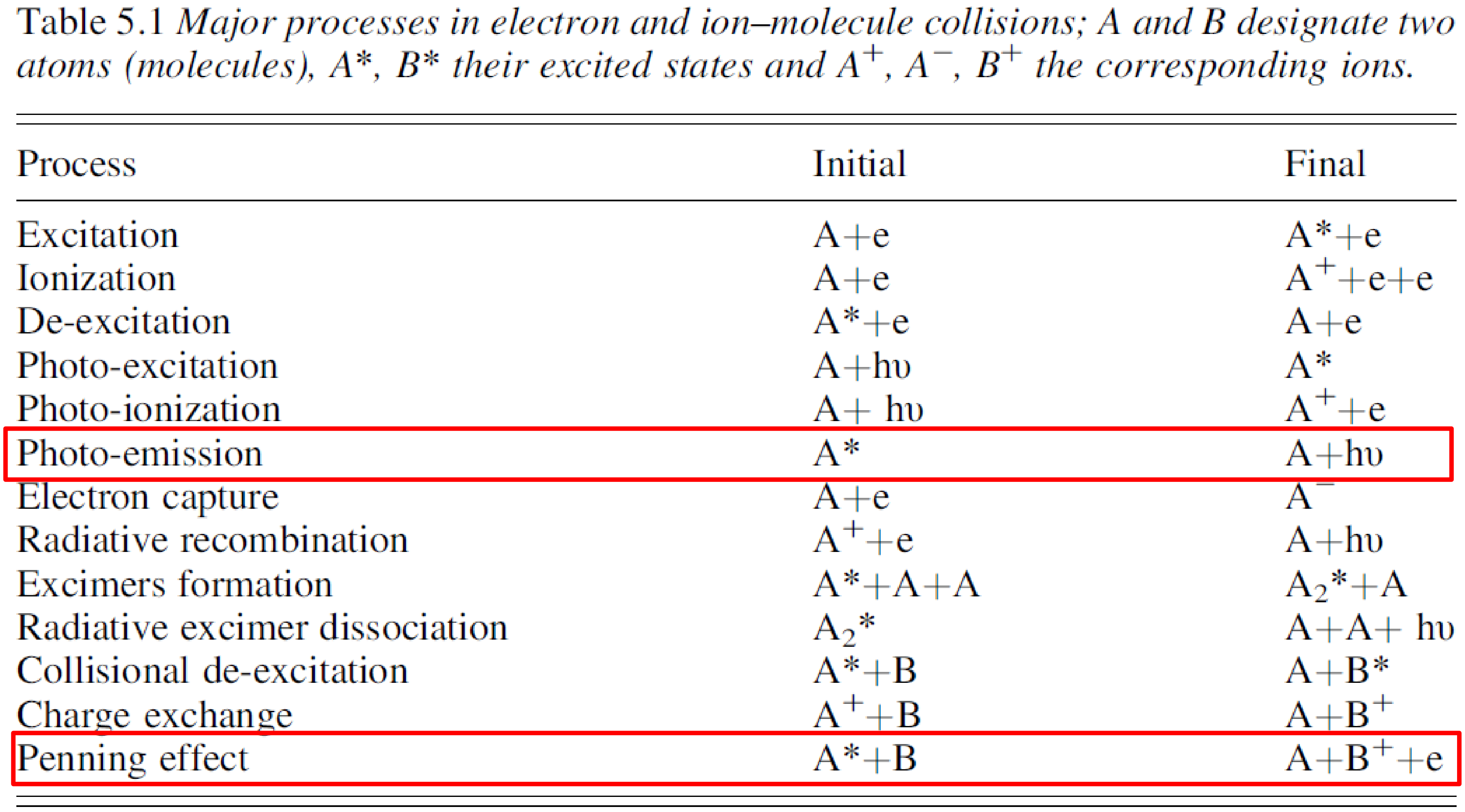


Table 5.1 *Major processes in electron and ion–molecule collisions; A and B designate two atoms (molecules), A^*, B^* their excited states and A^+, A^-, B^+ the corresponding ions.*

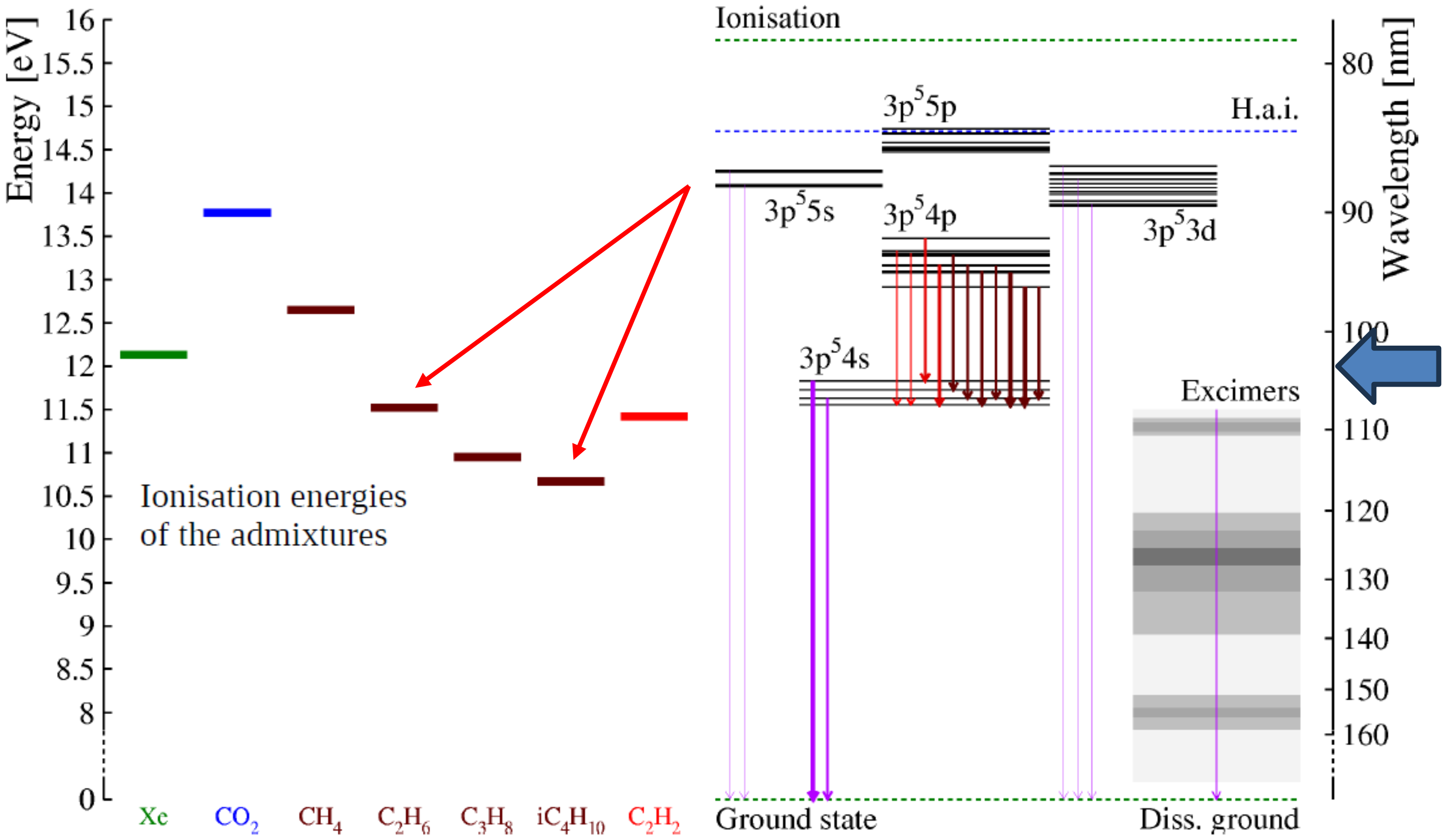
Process	Initial	Final
Excitation	$A+e$	A^*+e
Ionization	$A+e$	A^++e+e
De-excitation	A^*+e	$A+e$
Photo-excitation	$A+h\upsilon$	A^*
Photo-ionization	$A+ h\upsilon$	A^++e
Photo-emission	A^*	$A+h\upsilon$
Electron capture	$A+e$	A^-
Radiative recombination	A^++e	$A+h\upsilon$
Excimers formation	A^*+A+A	A_2^*+A
Radiative excimer dissociation	A_2^*	$A+A+ h\upsilon$
Collisional de-excitation	A^*+B	$A+B^*$
Charge exchange	A^++B	$A+B^+$
Penning effect	A^*+B	$A+B^++e$

From: F. Sauli, Gaseous radiation detectors: Fundamentals and Applications, Cambridge University Press, 2014

Penning effect

When using gas mixtures, Penning effect can take place, if the excitation potential on one species is larger than ionization potential of the other.

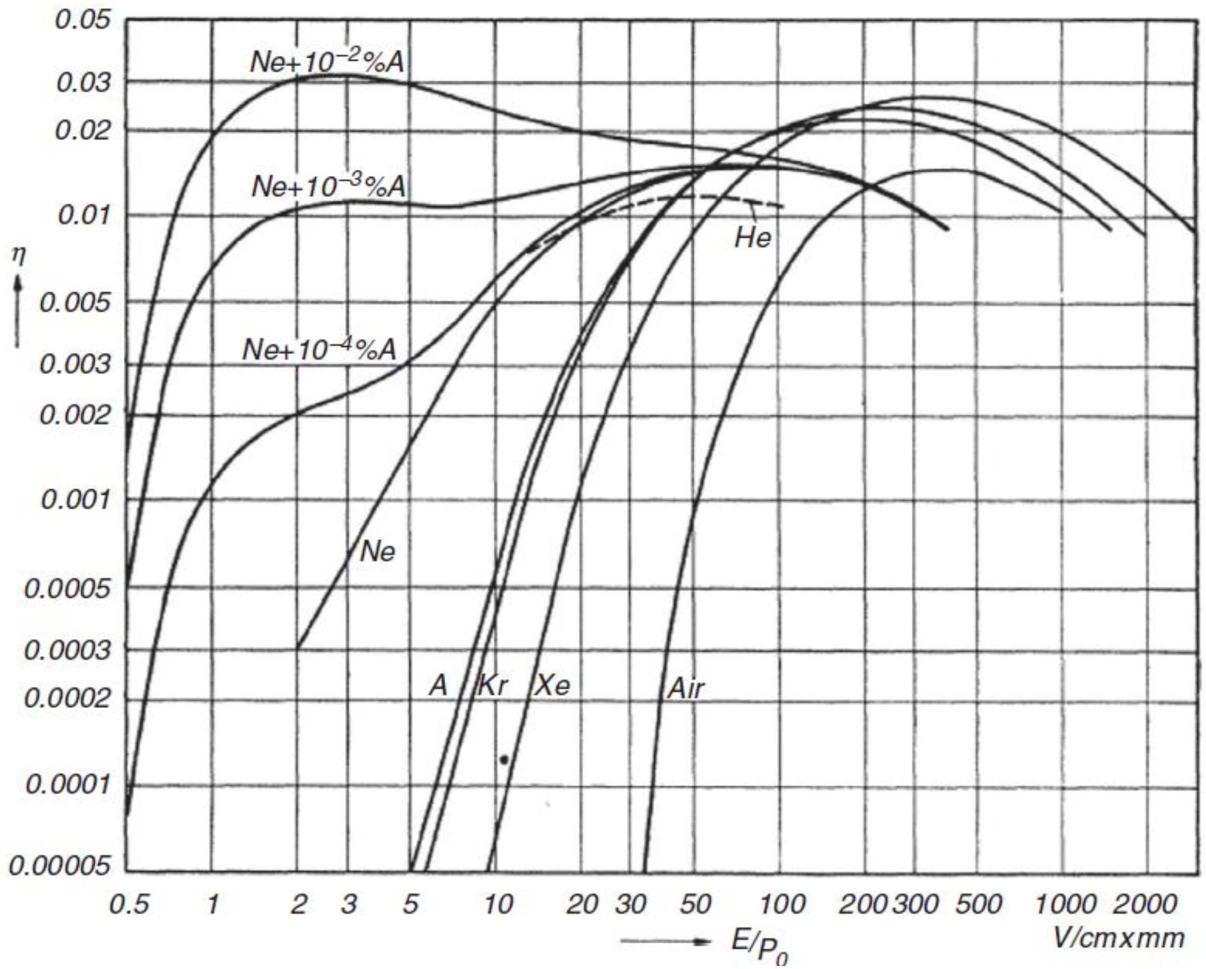
$$A^* + B \rightarrow A + B^+ + e$$



Ar excitation and ionization energy levels

Penning effect

Additional ion-electron pairs are produced, increasing the «effective» ionization coefficient, especially during multiplication processes.



From the original paper by Penning et al., 1940

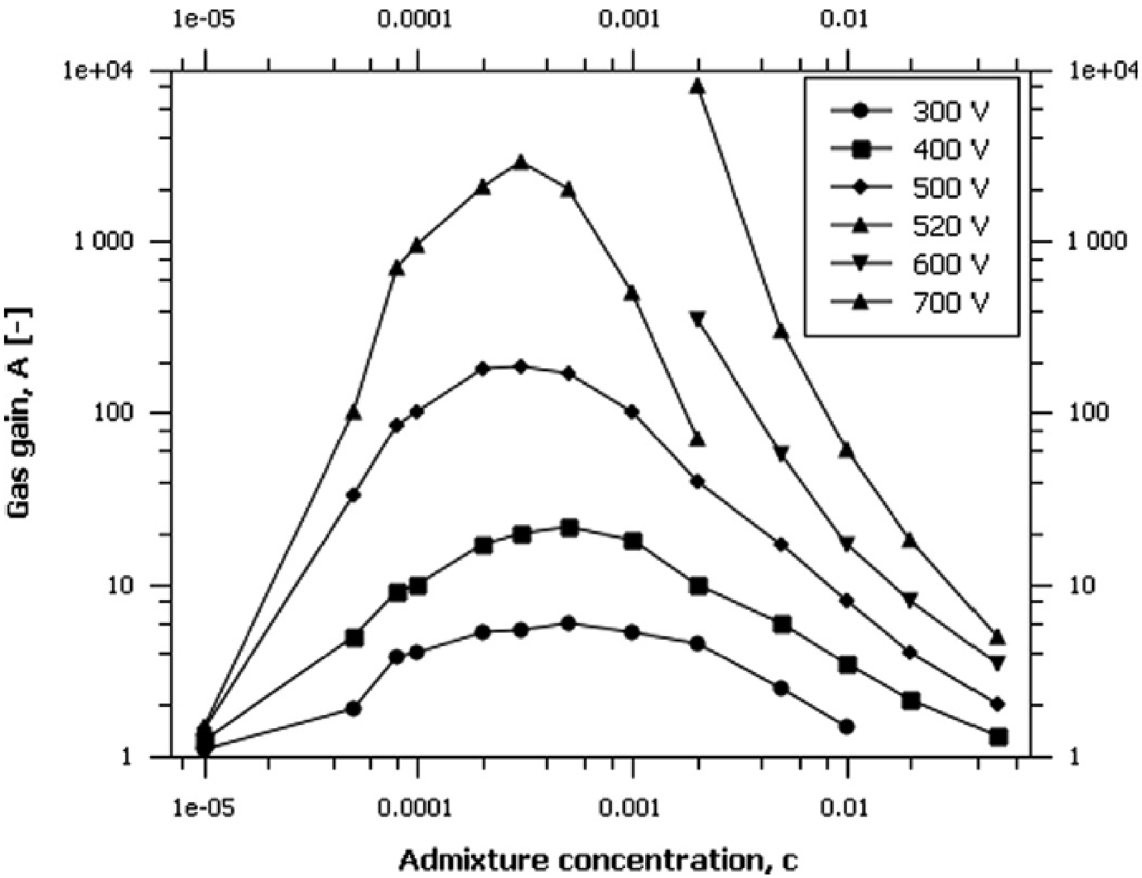
Ionization coefficient of Ne/Ar mixtures larger than ionization coefficients both of Ar and Ne

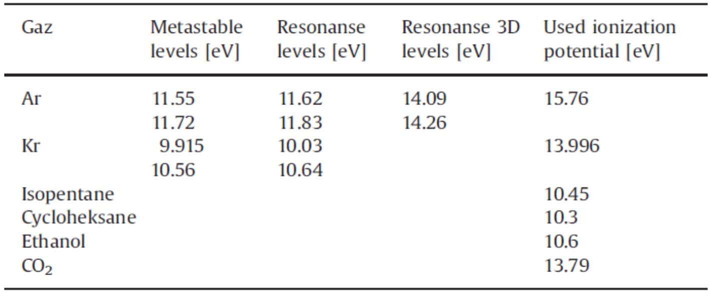
Penning effect: complications

Gain in an gaseous ionization counter, filled with Ar/isopentane mixture.

Two effects:

- Increase of the gain due to Penning effect for low concentration of iso-pentane
- Decrease of the gain for larger concentrations due to electron cooling by iso-pentane





Gaz	Metastable levels [eV]	Resonanse levels [eV]	Resonanse 3D levels [eV]	Used ionization potential [eV]
Ar	11.55 11.72	11.62 11.83	14.09 14.26	15.76
Kr	9.915 10.56	10.03 10.64		13.996
Isopentane				10.45
Cycloheksane				10.3
Ethanol				10.6
CO_2				13.79

http://dx.doi.org/10.1016/j.nima.2013.09.022

Choice of the gas filling

If only a noble gas is used to fill the counter, it can operate at relatively low gains ($M \approx 10^3 - 10^4$) before entering the permanent discharge regime

When the avalanche forms, excited Ar atoms are created, which can return to the ground state only emitting UV photons

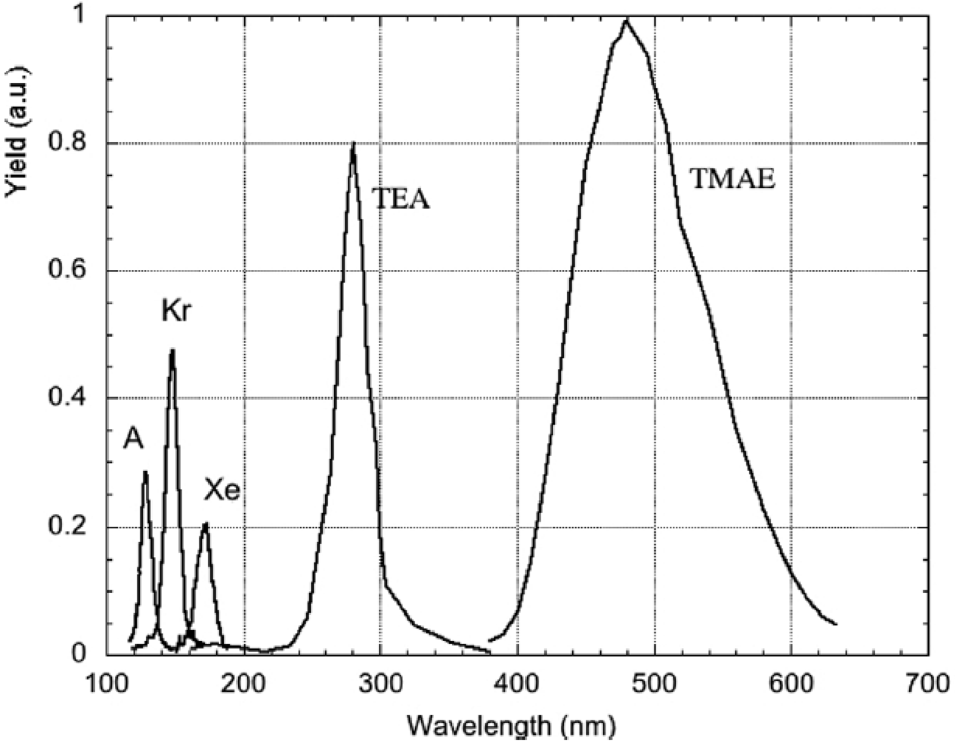
- The energy of these photons can be sometimes larger than the energy needed to extract electrons from the cathode walls (7.7 eV for copper)
- Photoelectrons can be extracted from the walls, which cause farther ionization of the gas

Possible spurious delayed pulses due to ions neutralizing on the cathode

- Ar+ ions reaching the cathode are neutralized
- The neutral Ar atom is sometimes produced in an excited state, decays emitting UV photons → An additional electron can also be extracted from the wall

Both processes induce spurious delayed pulses

Emission spectra from various gases



Emission spectra of the noble gases typically fall in the UV region

Figure 5.7 Luminescence emission in argon–xenon mixtures (Takahashi *et al.*, 1983). By kind permission of Elsevier.

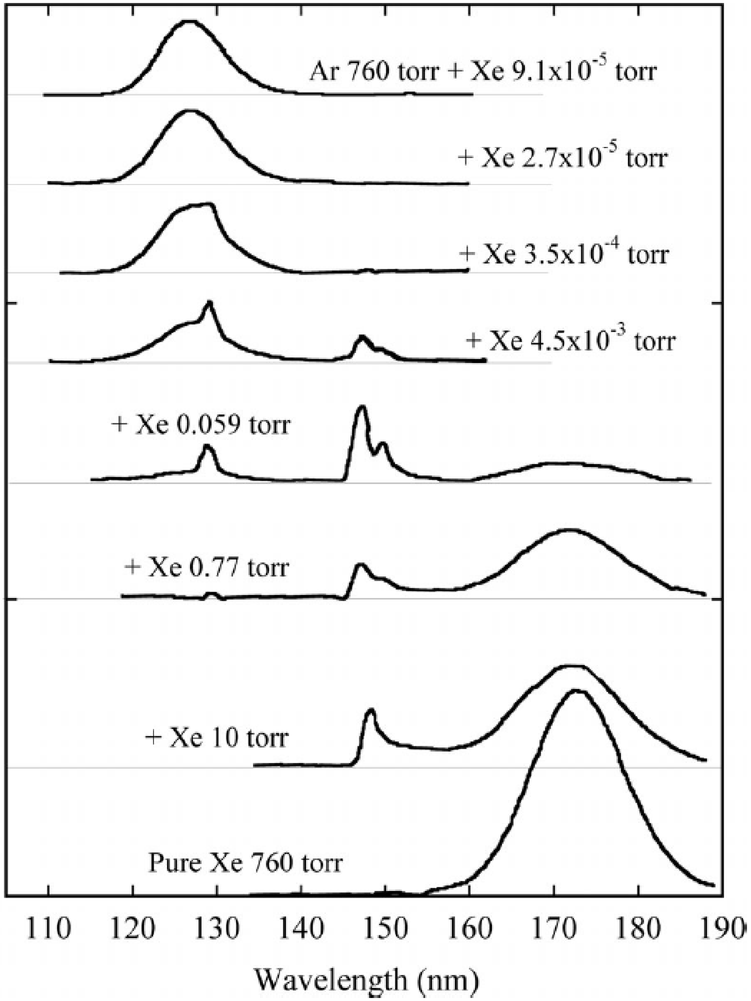


Figure 5.12 Secondary emission spectra of A, Kr, Xe, TEA and TMAE (Breski *et al.*, 1988). By kind permission of Elsevier.

Radiation trapping

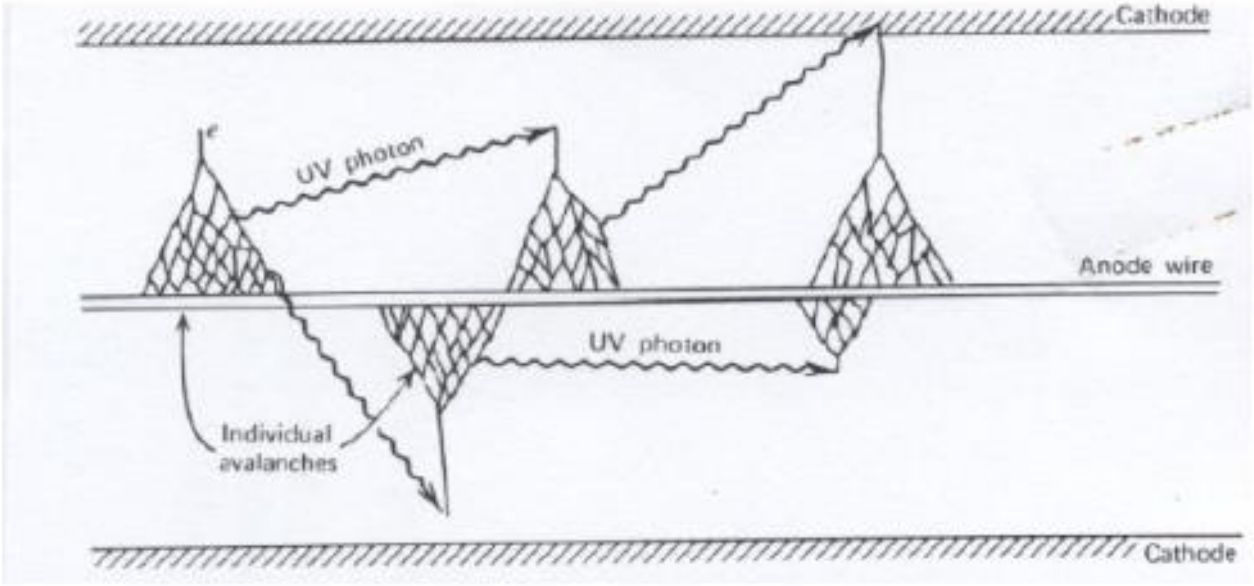
- Photons emitted by radiative decay to ground of excited noble gas atoms have a high probability of being re-absorbed and subsequently re-emitted by the noble gas: radiation trapping.

- Eventually, after many absorption/emission cycles, the photons are absorbed by the walls of the gaseous detector (or a quencher gas molecule).

- Excited states may effectively have a long lifetime

Geiger-Muller counters

Filling gases: He, Ne or Ar
High electric fields
Gains of the o(10^{10})



Entire gas volume is involved
→ Loss of information on space
→ Loss of information on energy
Simple, easy to use, cheap, suitable for measuring radiation counts
Sometimes optimization of gas mixtures is easy… ☺

A Geiger-Muller counter built in 1939 and used in the 1947-1950 for cosmic ray studies in balloons and on board B29 aircraft by Robert Millikan et al.

Made of copper, 30 cm long

Use of quencher gases

The problem of UV photons is solved by adding to noble gas percentages of other polyatomic gases, usually called "quenchers"

✓Gas quencher molecules have non-radiative (rotational and vibrational) mode and can absorb UV photons in a wide energy range.

- CH_4 absorbs photons between 7.9 and 14.5 eV
- Other polyatomic gases used are freons, CO_2, BF_3, C_4H_{10}, etc.

✓Polyatomic gases dissipate energy by elastic collisions

- Some of these gases can dissociate, and/or form solid or liquid polymers which can deposit on the electrodes, altering their properties

→Malter effect, where the E field close to the electrode is distorted, causing undesired discharges

→Aging effects (one of the main issues in all particle detectors)

Even small quantities of quencher allow to operate these detectors up to gain of 10^6 without discharges.

Quenching by UV absorption

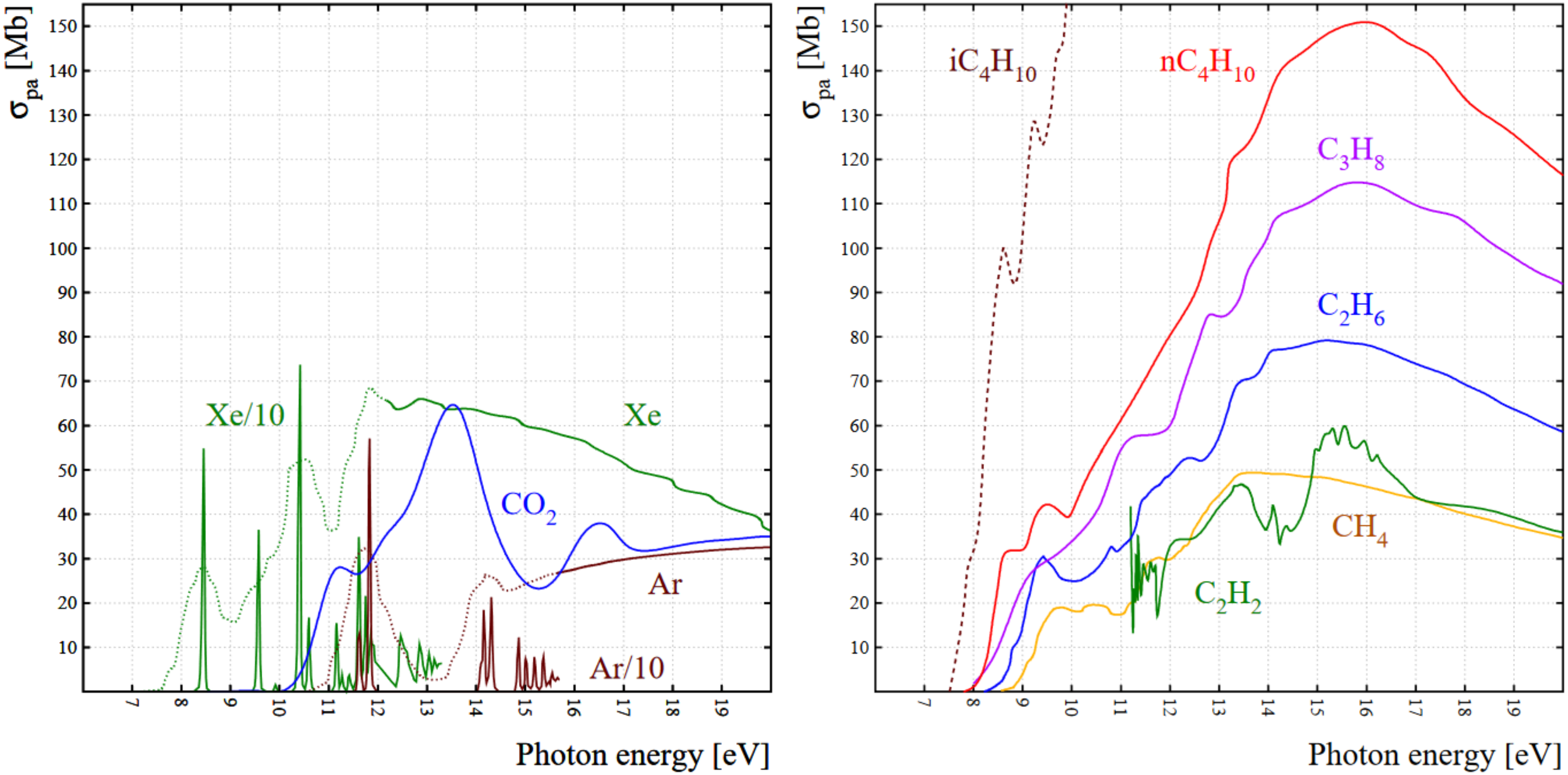


Photo-absorption cross sections for various gases and for the principal hydrocarbons

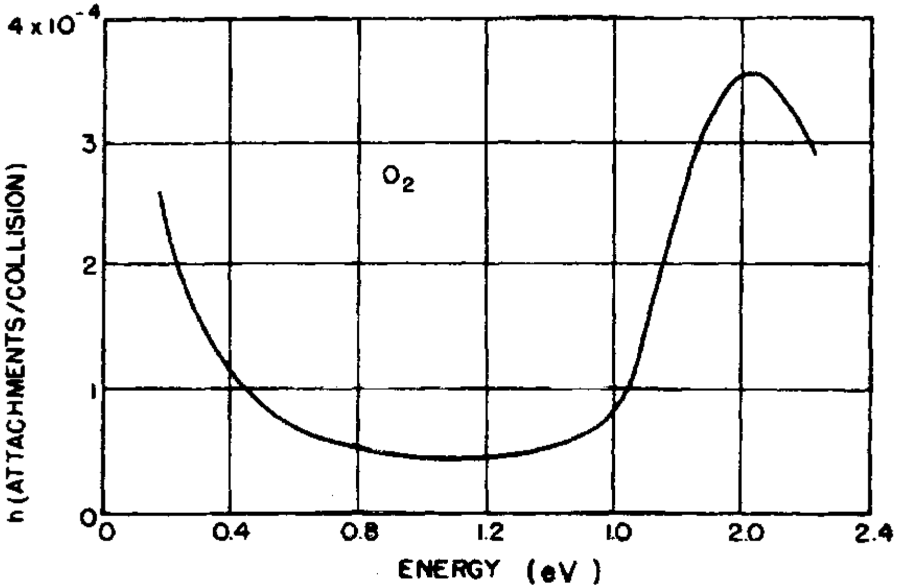
→ They extend down to 7-8 eV

Electronegative gases

- Some gases, like O_2, Cl_2, H_2O vapour, etc., are characterized by a high probability to capture drifting electrons and form negative ions;
- The net effect is reducing the total number of drifting electrons → electronegative gases
- The probability of capture per collision (or per unit length) is called "attachment coefficient"

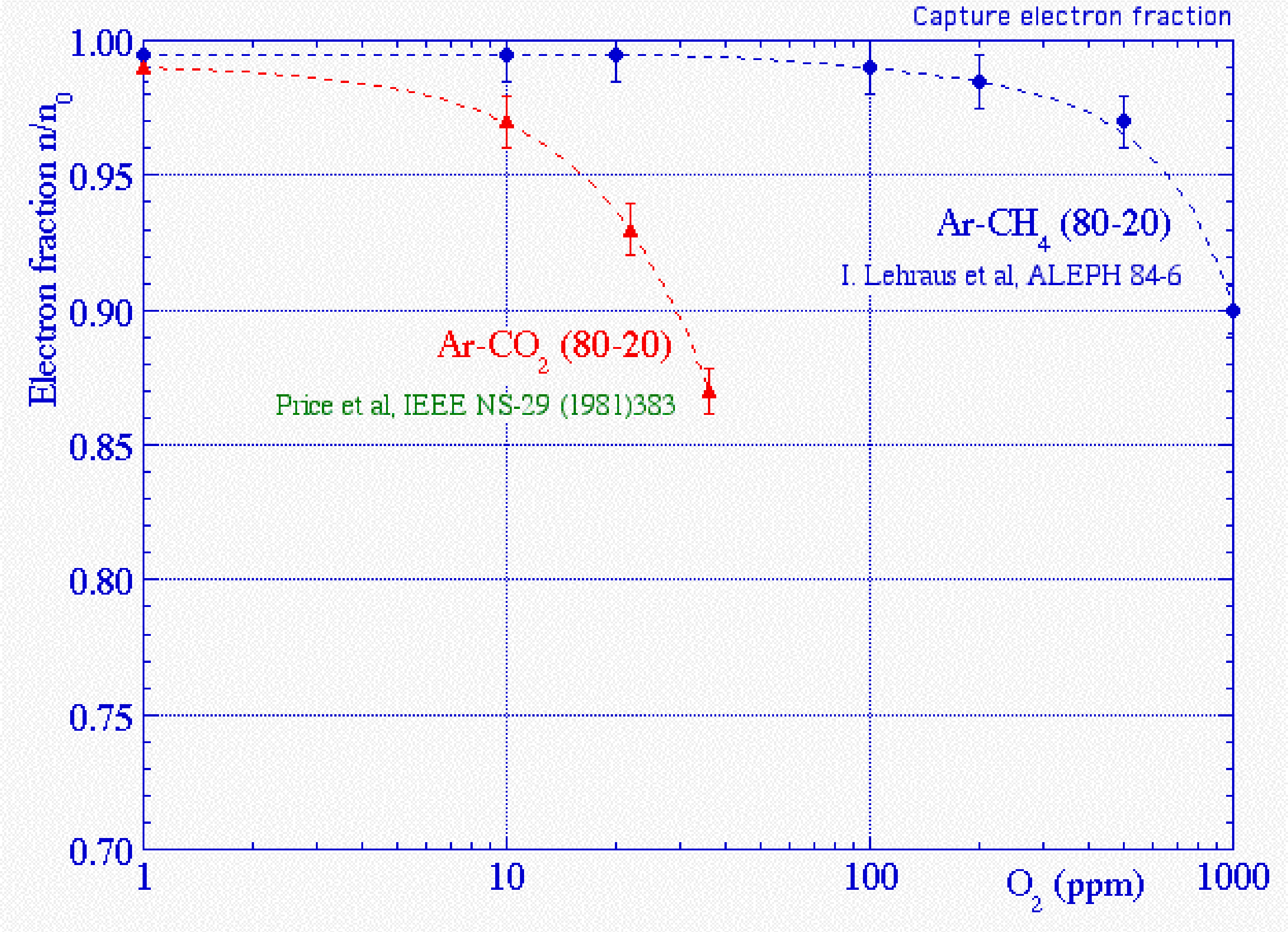
The attachment coefficient strongly depends on electron energy .

Attachment coefficient of electrons in Oxigen, vs. electron kinetic energy



Effects of electronegative gases

Fraction of electrons surviving after a drift of 20 cm in two gas mixtures, as a function of the contamination of O_2, under the action of E = 200 V/cm



Use of quencher gases

Also electronegative gases are used as quenchers, like many freons (CF_3Br), ethyl-bromide (C_2H_5Br), etc. which help operate detectors at high gain without discharges.

- They are characterized by an electron capture mean free path that is less than typical anode-cathode distance (≈ 1 cm) and larger than avalanche size (few μm)

- These gases capture electrons possibly extracted at the cathode forming negative ions, preventing the formation of spurios delayed avalanches

- Sometimes quenchers, in particular various kinds of freon, can dissociate resulting in simpler molecules, sometimes harmful to the detector, like HF → aging

«Classical» gas mixtures

Popular gas mixtures in use for a wide variety of gaseous detectors were ternary:

- Noble gases (typically Ar) to enhance primary, secondary ionization and Towsend avalanches in the gas
  → enhance the signal produced
  → Reduce the intrisic inefficiency
  → They do not dissociate in pollutants
- Small fractions of hydrocarbons (CH_4 or i-C_4H_{10}), or CO_2, and/or fractions of some kind of Freon

- When no simulation were available the «best» gas mixture for a certain detector was chosen by making, literally, hundreds of tests
- Simulations have made life easier to particle detector physicists

Quenching gas for detectors of charged particles

PATENT · 22 gennaio 1974

OSTI ID: 4339272

Atac, M

Operation of detectors of charged particles such as wire counters and Geiger-Muller tubes is improved by filling the counters with a quenching-gas mixture of argon, isobutane and methylchloroform. (Official Gazette)

Search for the full text at the U.S. Patent and Trademark Office

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Details Similar Records / Subjects

Research Organization:	Originating Research Org. not identified
NSA Number:	NSA-29-026886
Assignee:	to United States Atomic Energy Commission
Patent Number(s):	US 3787746
OSTI ID:	4339272
Resource Relation:	Patent File Date: 1973 May 11; Other Information: H01j39/26. Orig. Receipt Date: 30-JUN-74
Country of Publication:	United States
Language:	English

Similar Records

Feasibility of using electronic quenching to eliminate halogen gas in Geiger--Muller detectors in aircraft oil gauging systems
Technical Report · 1975 · OSTI ID: 4339272
McClellan, J L

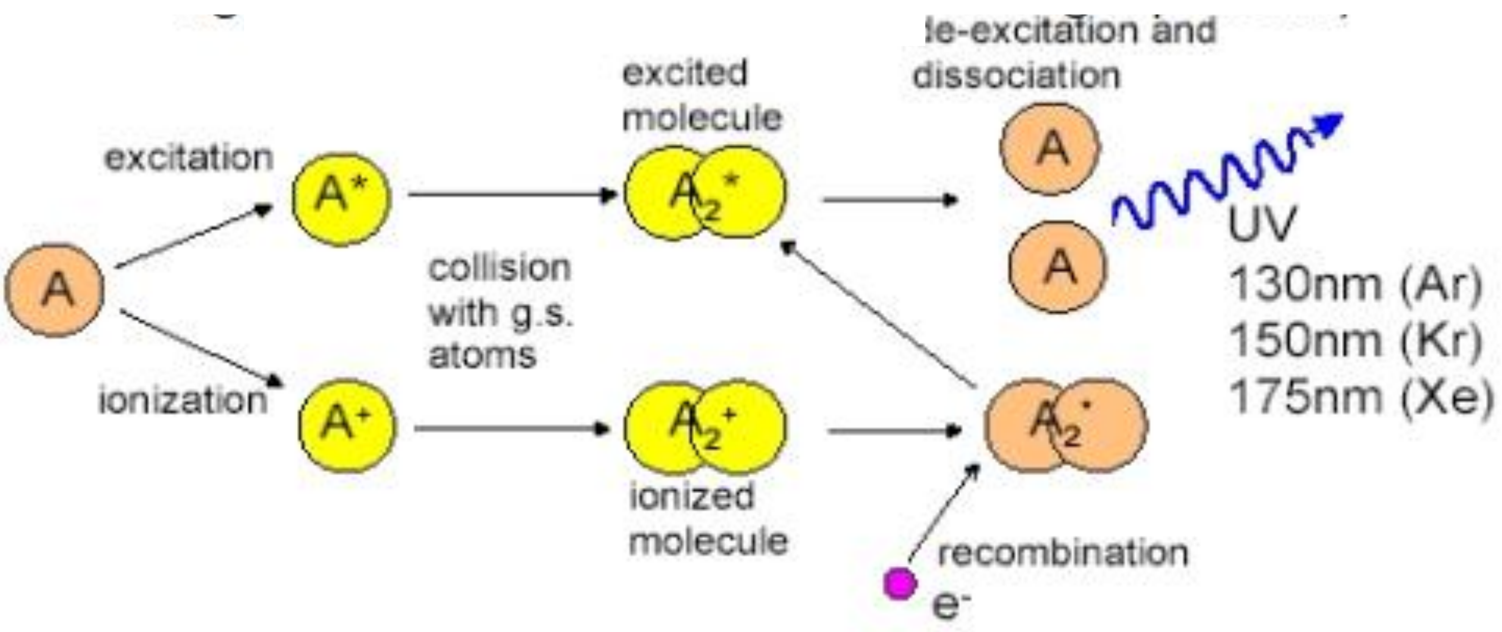
LIGHT PULSE FROM HALOGEN-QUENCHED GM COUNTER DISCHARGES
Journal Article · 1962 · Journal of Applied Physics (U.S.) · OSTI ID: 4339272
Davis, Jr, W P; Worthington, W C

Method and apparatus for measuring subchannel voids in a light water reactor test fuel assembly
Patent · 1992 · OSTI ID: 4339272
Shiraishi, L M; Wilhelmson, D A; Matzner, B

Related Subjects

- N46110* -Instrumentation-Radiation Detection Instruments- General Detectors & Monitors
- *GEIGER-MUELLER COUNTERS
- 2-METHYLPROPANE CHLORINATED ALIPHATIC HYDROCARBONS
- ARGON
- CHLORINATED ALIPHATIC HYDROCARBONS
- GASES
- MIXTURES
- ORGANIC CHLORINE COMPOUNDS
- QUENCHING

Exploiting ligth emission



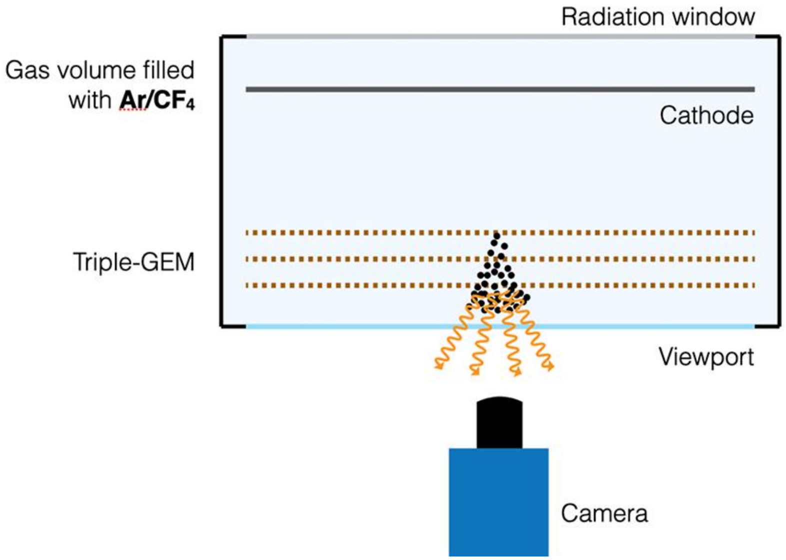
- ✓ Charged particles leave trails of excited molecules
- ✓ Certain gases release part of this excitation energy producing visible or UV photons
- ✓ This can be exploited if the gases are transparent to these photons

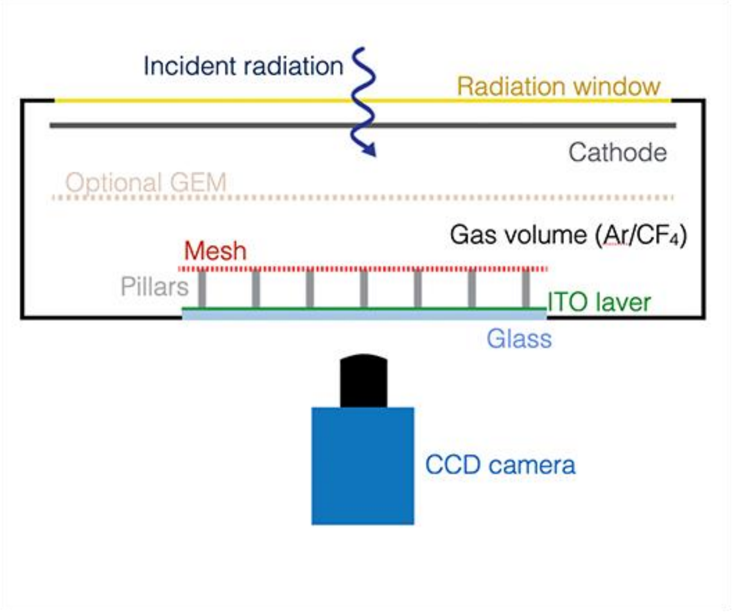
Gaseous detectors with optical readout

Energy carried away by scintillation light is small: < 1% of dE/dx

- Up to 10^4 photons per MeV of dE/dx

  → use the scintillation ligtht produced during the avalanche multiplication processes

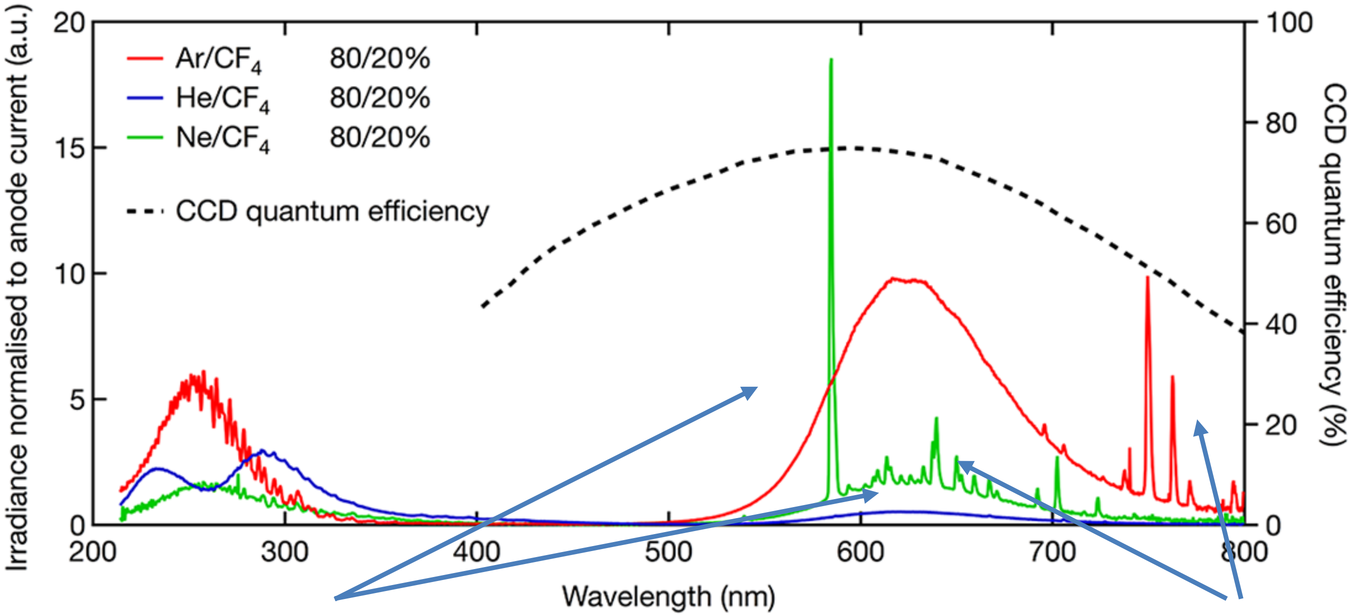




Typically Micro Pattern Gaseous Detectors (MPGDs) are used coupled with CCD or CMOS cameras.

Exploiting ligth emission

Spectrum of the noble gases is typically in the UV region.
Maximum sensitivity of photosensors is typically for visible light.
→ Need to use wavelength shifters
→ Add suitable gases to noble gases

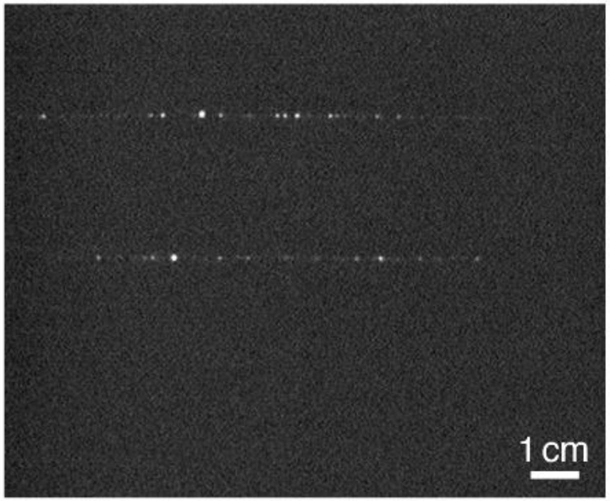


Broad spectrum from CF_4

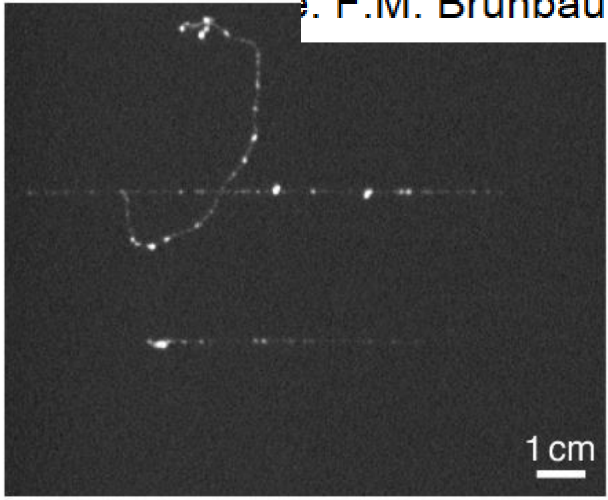
Sharp peaks from noble gases

Images from optical readout GDs

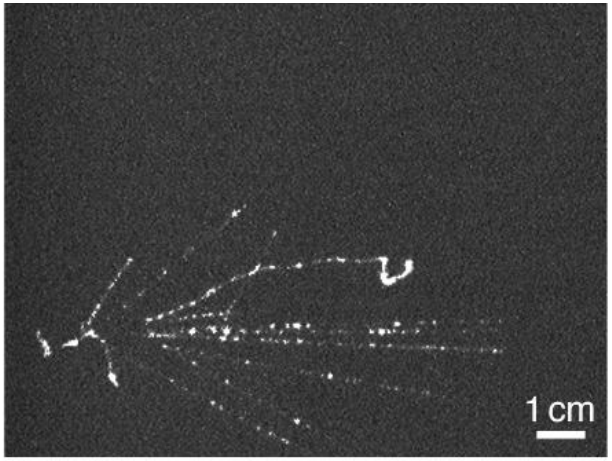
: F.M. Brunbauer *et al* 2018 *JINST* **13** T02006



Muon tracks



Muon + δ ray



Hadronic shower

Scintillation light decay time:

- ✓ ≈ 10 μs for noble gases → suitable for typical CCD integration time
- ✓ 10-1000 ns in inorganic scintillatos
- ✓ 2 ns for fast organic scintillator

First particle detectors were «imaging» detectors

→ Intuitive and direct visualization of what happens

Electron Drift velocity

Ionization probabilty (and gain) are not the only parameters to be optimized in gaseous detectors.

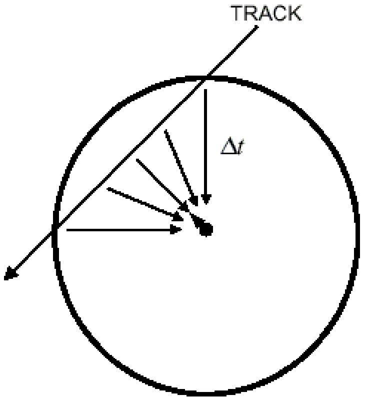
✓ Drift velocity is typically related to time resolution in GDs.

- In RPC, where the whole volume is active for multiplication processes:

$$\boxed{\sigma_t} = \sqrt{\sigma^2_{n_{cl}} + \sigma^2_{n_0} + \sigma^2_M} = \frac{1}{\boxed{v_d}} \frac{1}{\eta - \frac{1}{g}} \sqrt{\left(\frac{\sigma_{n_{cl}}}{n_{cl}}\right)^2 + \left(\frac{\sigma_{n_0}}{n_0}\right)^2 + \left(\frac{\sigma_M}{M}\right)^2}$$

In wire chambers and most MPGDs time resolution is related to fluctuations in the drift time toward th amplification region.

- CF_4 is added to Ar to improve time resolution



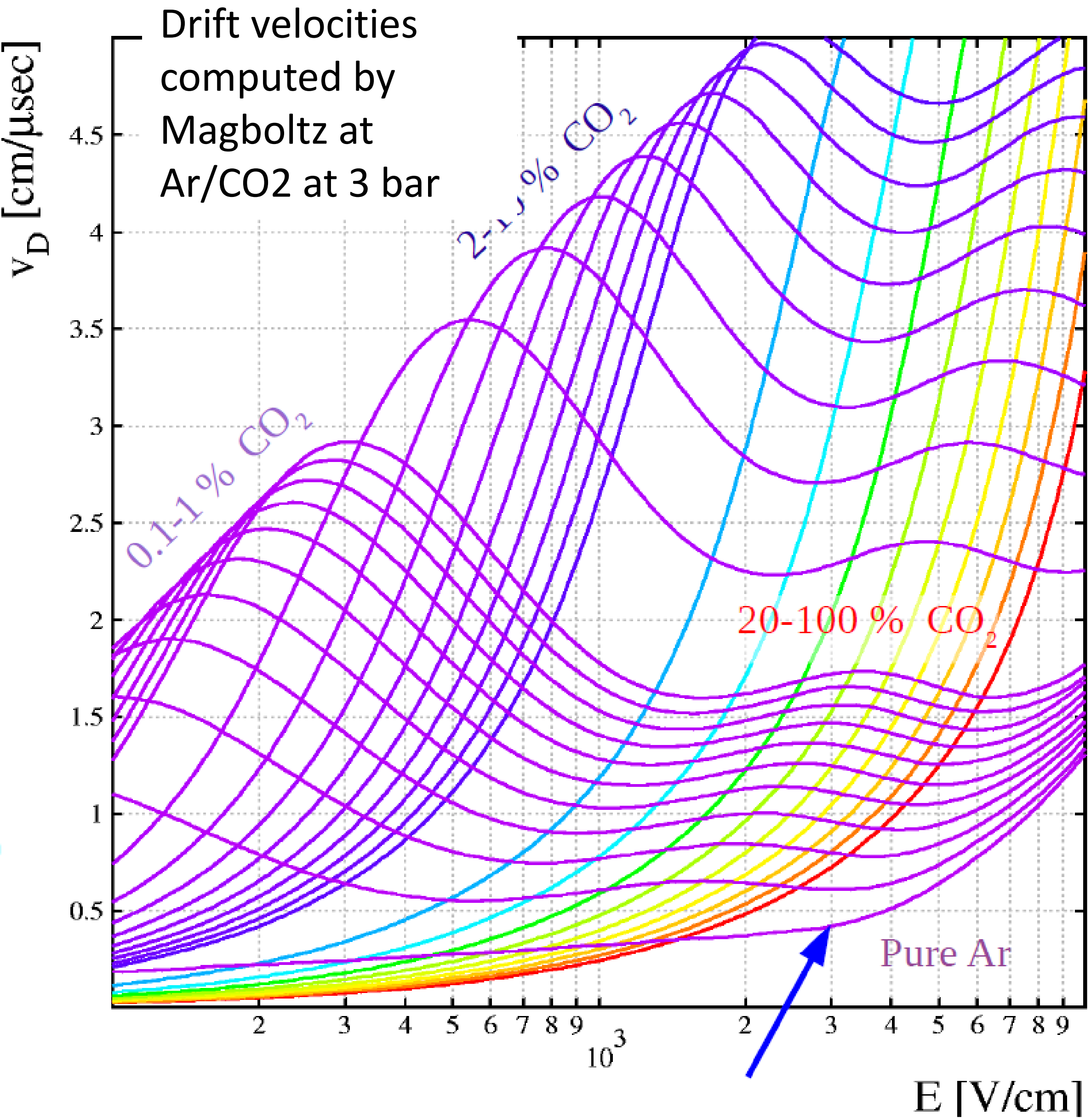
In general:

$I_{ind} = q_e v_d E_w$ → the signal amplitude is proportional to drift velocity

Drift velocity in Ar/CO_2

- Adding even small percentages of a gas to another can change the drift velocity dramatically
- CO_2 added to Ar makes the drift velocity of the mixture max larger

- In general, mixtures characterized by large drift velocities are preferred: «fast» gas mixtures

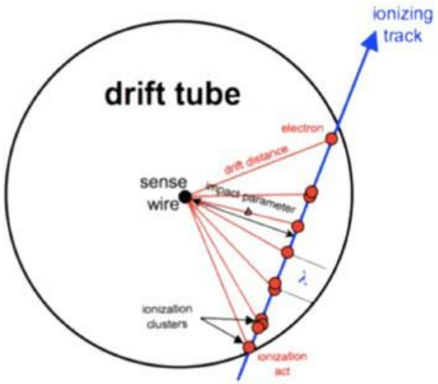


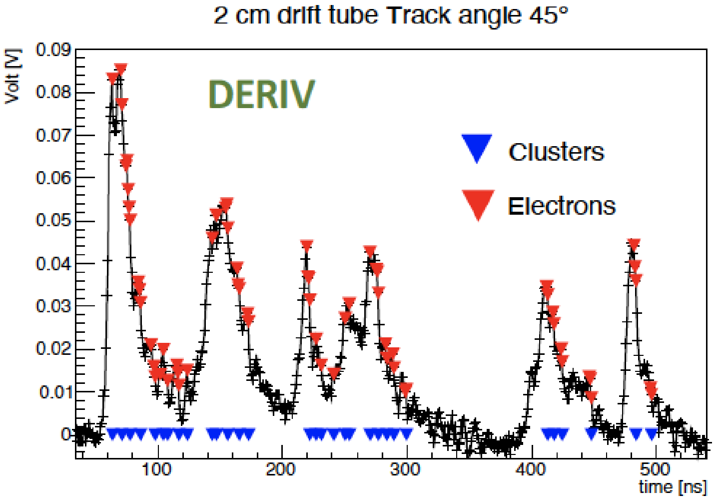
Drift velocities computed by Magboltz at Ar/CO2 at 3 bar

«Slow» gas mixtures: the cluster counting technique

Principle: In He based gas mixtures the signals from each ionization act can be spread in time to few ns.

- By counting the number of ionization acts per unit length (dN/dx), it is possible to identify the particles (P.Id.) with a better resolution w.r.t the dE/dx method.





- Landau distribution of dE/dx, originated by the mixing of primary and secondary ionizations, has large fluctuations and limits separation power of PID → primary ionization is a Poisson process, has small fluctuations

dE/dx: truncated mean cut (70-80%), with a 2m track at 1 atm give σ ≈ 4.3%

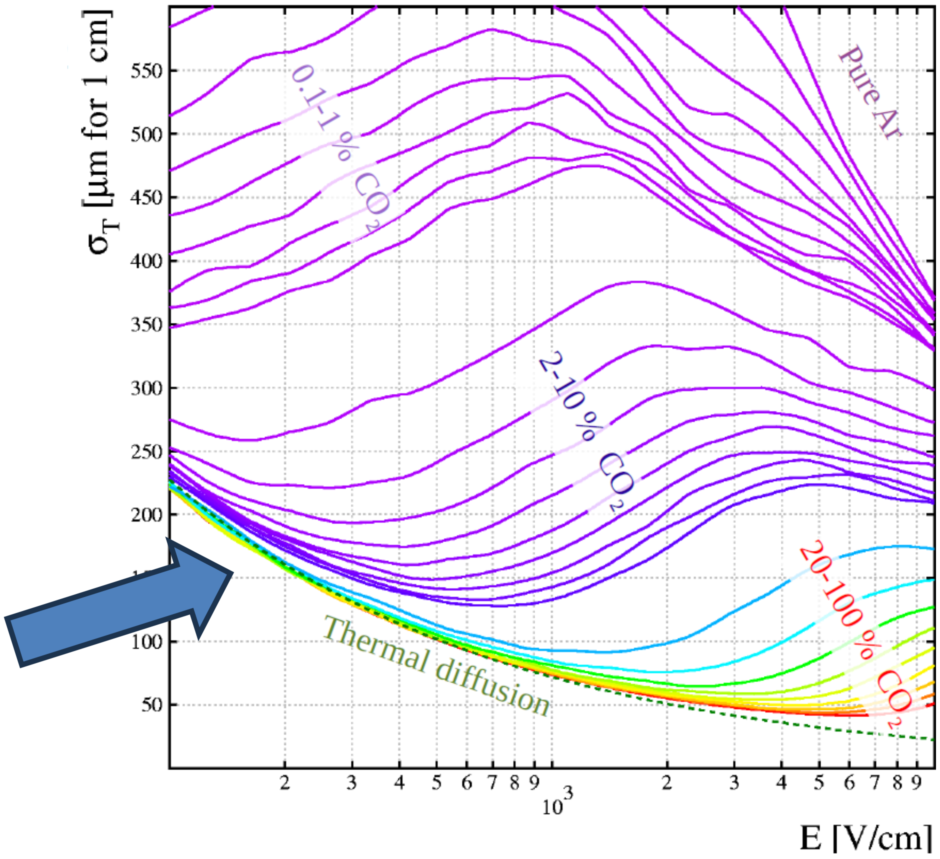
dN_{cl}/dx: for He/iC_4H_{10}=90/10 and a 2m track gives $\sigma_{dNcl/dx}$ **/(dN_{cl}/dx)** < 2.0%

Diffusion coefficients for Ar/CO_2

Transversal diffusion related, typically, to spatial resolution
Longitudinal diffusione related, sometimes, to time resolution
→ In drift tube (or TPC) it can affect spatial resolution as well

- Transversal and longitudinal diffusion should be optimized together with other parameters (gain, drift velocity).

- Calculated by Magbolts for Ar/CO_2 at 3 bar



Diffusion of electrons and ions in gases

While diffusing, electrons and ions do not have an easy life.

Possible interaction of positive ions:

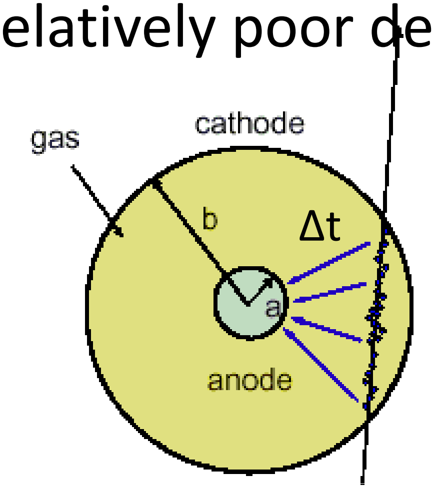
- Neutralization with a negative charge carrier (either an electron or a negative ion)
- Neutralization after the extraction of an electron from the walls of the detector
- Charge transfer
  - to a molecule of its own gas ($A^+ + A \dashrightarrow A + A^+$)
  - to a molecule of another gas with lower ionization potential ($A^+ + B \dashrightarrow A + B^+$)

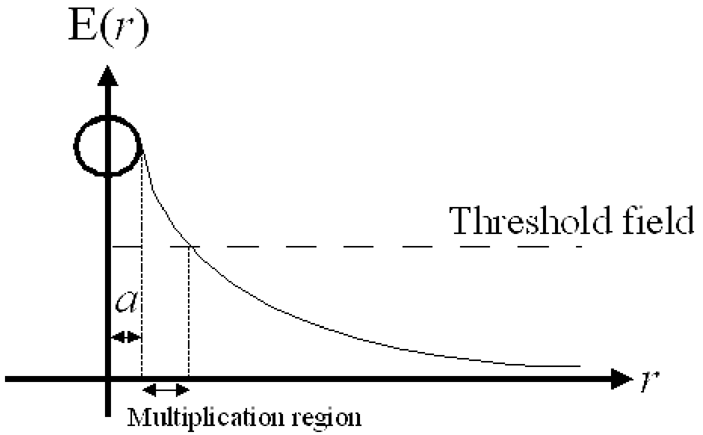
Possible interactions of electrons:

- Neutralization with a positive ion (e + A+ --> A)
- Attachment to a molecule with negative electron affinity (e + A --> A-)
  - This attachment probability is negligible for noble gases and hydrogen
  - Oxigen is a gas with strong electronegative affinity
  - The attachment coefficient depends strong on the electric field (if any)
- Absorption in the walls of the detector

Changing of paradygm

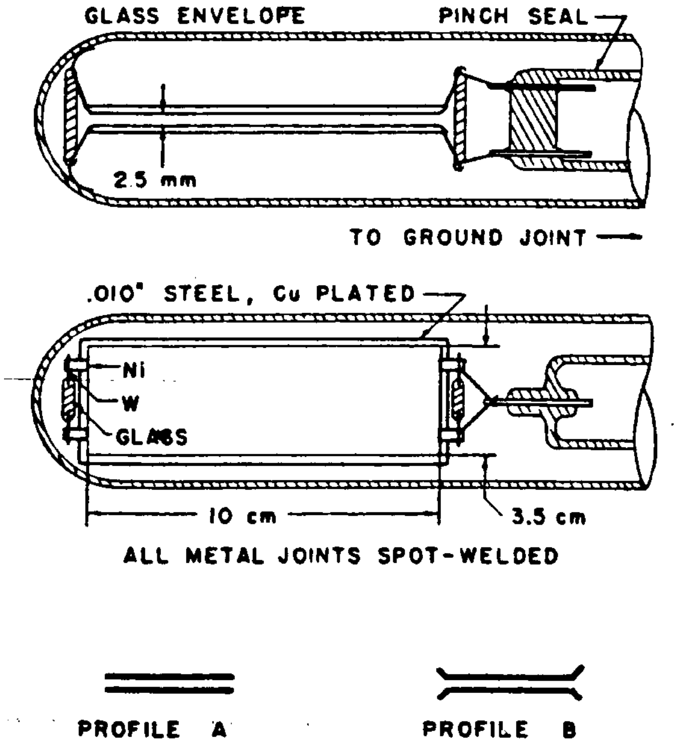
- Until 1980s most succesfully gaseous detectors were cylindrical
  - → yes, MWPCs are «cylindrical»
- Drift and multiplication region separated from each other
  - → quite good spatial resolution
  - → relatively poor devices for time resolution

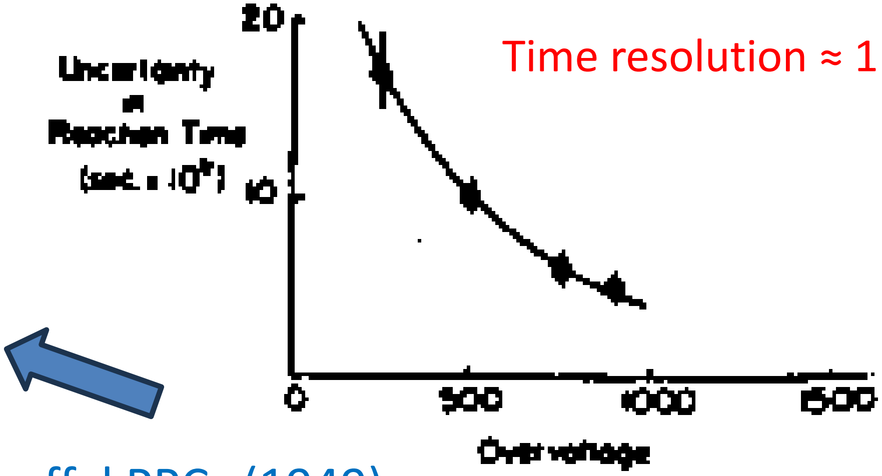




- Using a parallel plate geometry was considered difficult
  - →Detectors unstable and with limited lifetime
- Once that a discharge started the whole energy stored in the electrodes was dissipated
- Detectors had to be externally triggered

Changing of paradygm

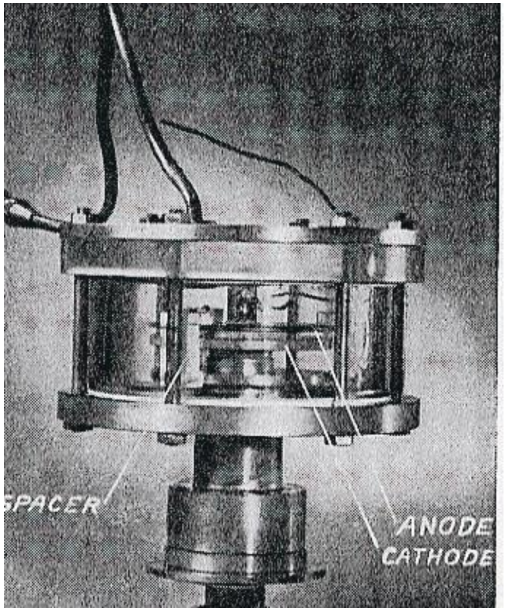




Time resolution ≈ 1 ns

Keuffel PPCs (1949)

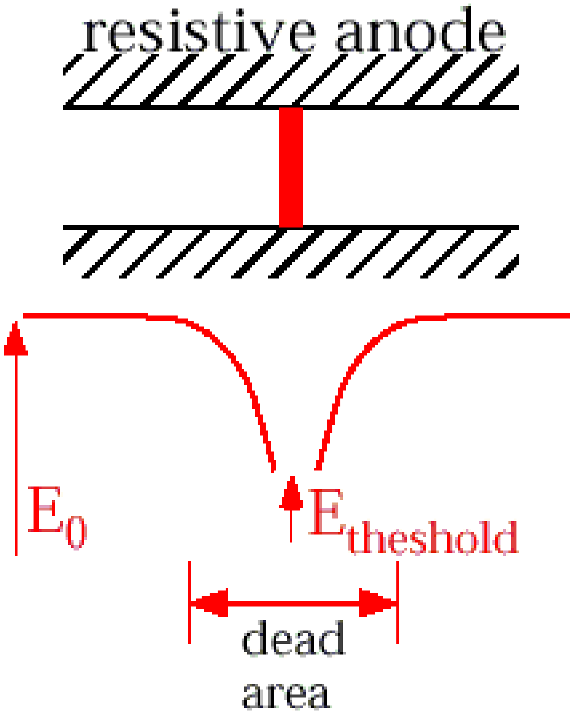
Madansky and PIDD PPCs

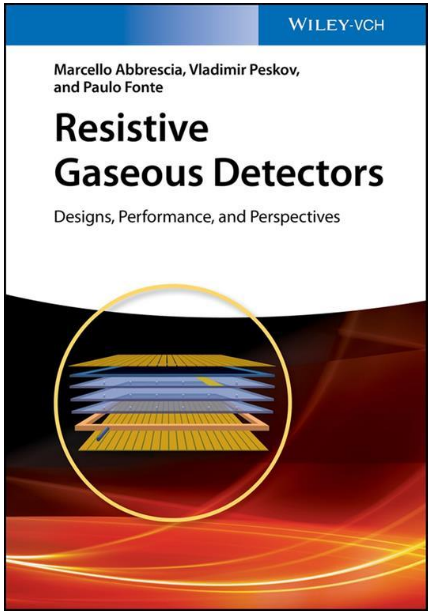


Including resistive elements in gaseous detectors

- Resistive electrodes allow auto-quenching mechanisms due to the switching off of the LOCAL electric field
- Limitation of rate capability

Including resistive elements in gaseous detectors was (and is) a major breakthrough in GD R&D




WILEY-VCH

Marcello Abbrescia, Vladimir Peskov, and Paulo Fonte

Resistive Gaseous Detectors

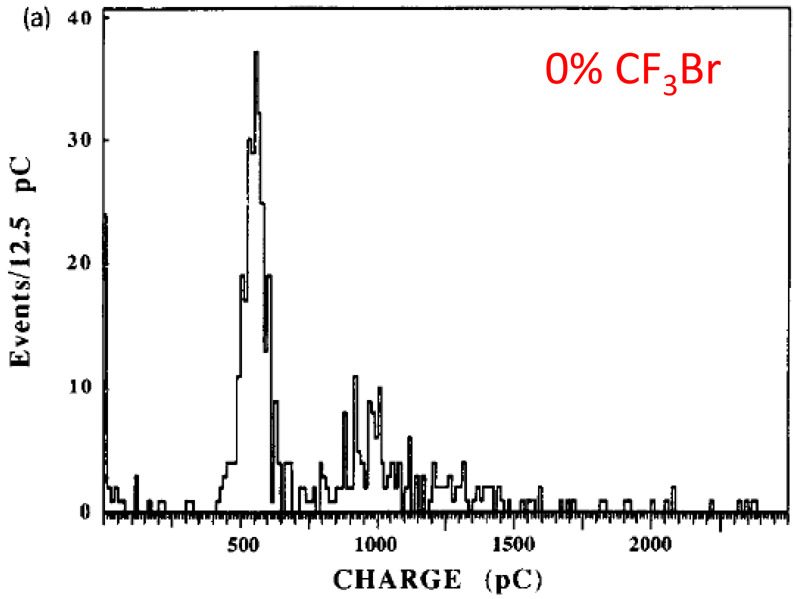
Designs, Performance, and Perspectives

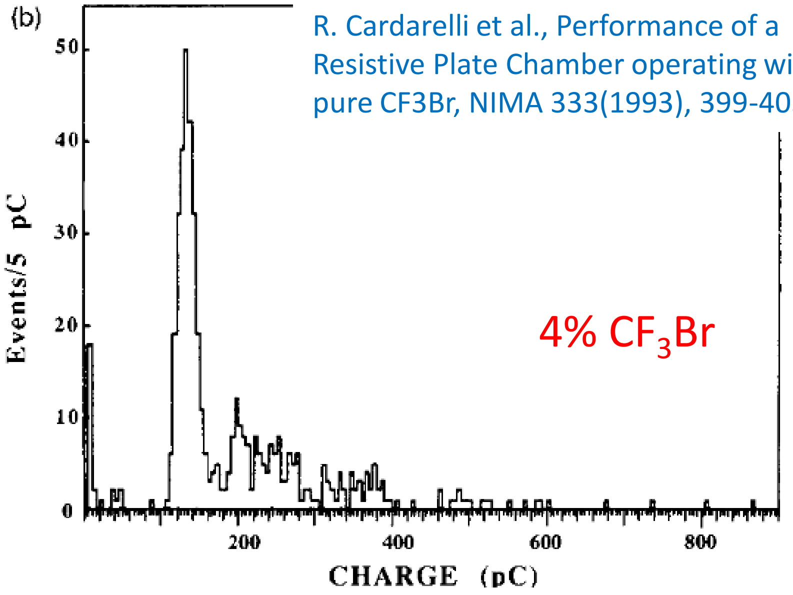

WILEY

阻性板
气体探测器
设计、性能及应用

RPCs succesfully operated with $Ar/iC_4H_{10}/CF_3Br$ mixtures

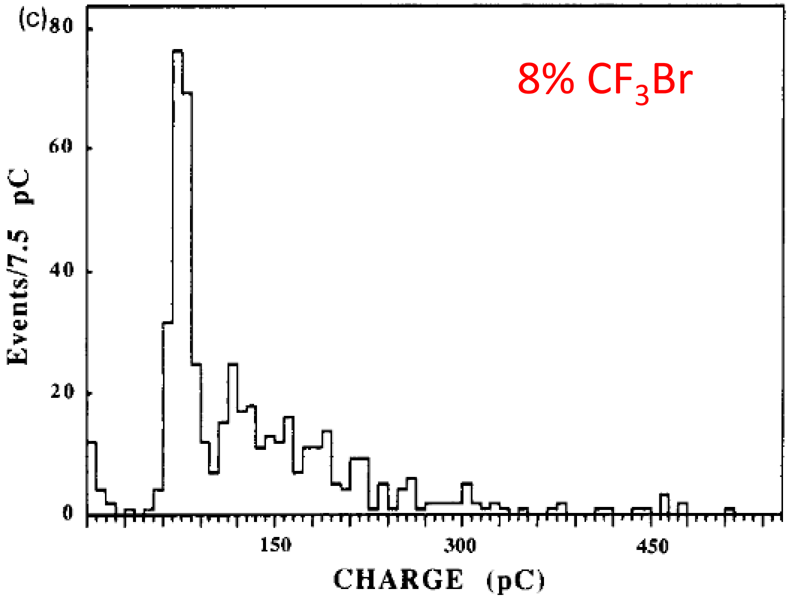
Trasferring amplification to front-end electronics





R. Cardarelli et al., Performance of a Resistive Plate Chamber operating with pure CF3Br, NIMA 333(1993), 399-403

- Sometime large gains (charge) are not acceptable:
- RPCs operated in streamer mode were NOT suitable for LHC experiments because of their too low rate capability
- The solution was to increase the fraction of quencher gases and transfer part of the needed amplification to the front-end electronics

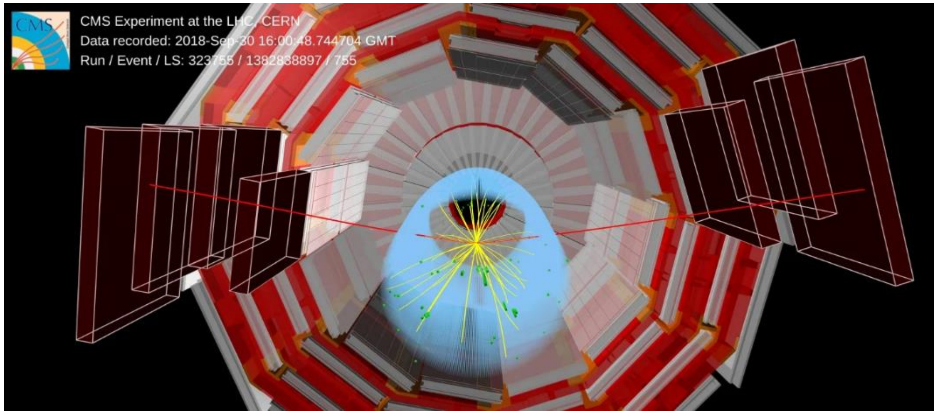


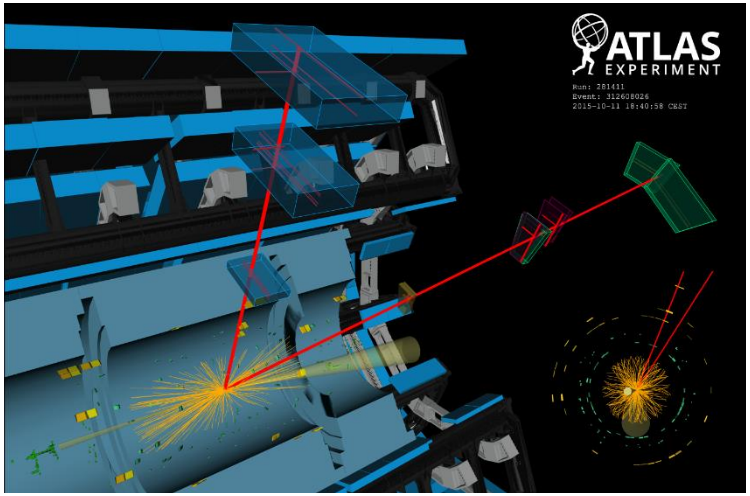
Passage from streamer to avalanche mode in RPCs

The use of CF_3Br enriched mixtures allowed an increase of the rate capability of more than one order of magnitude

→Allowed the use of RPCs in muon systems of the LHC experiments

→How a gas mixture can change your life…

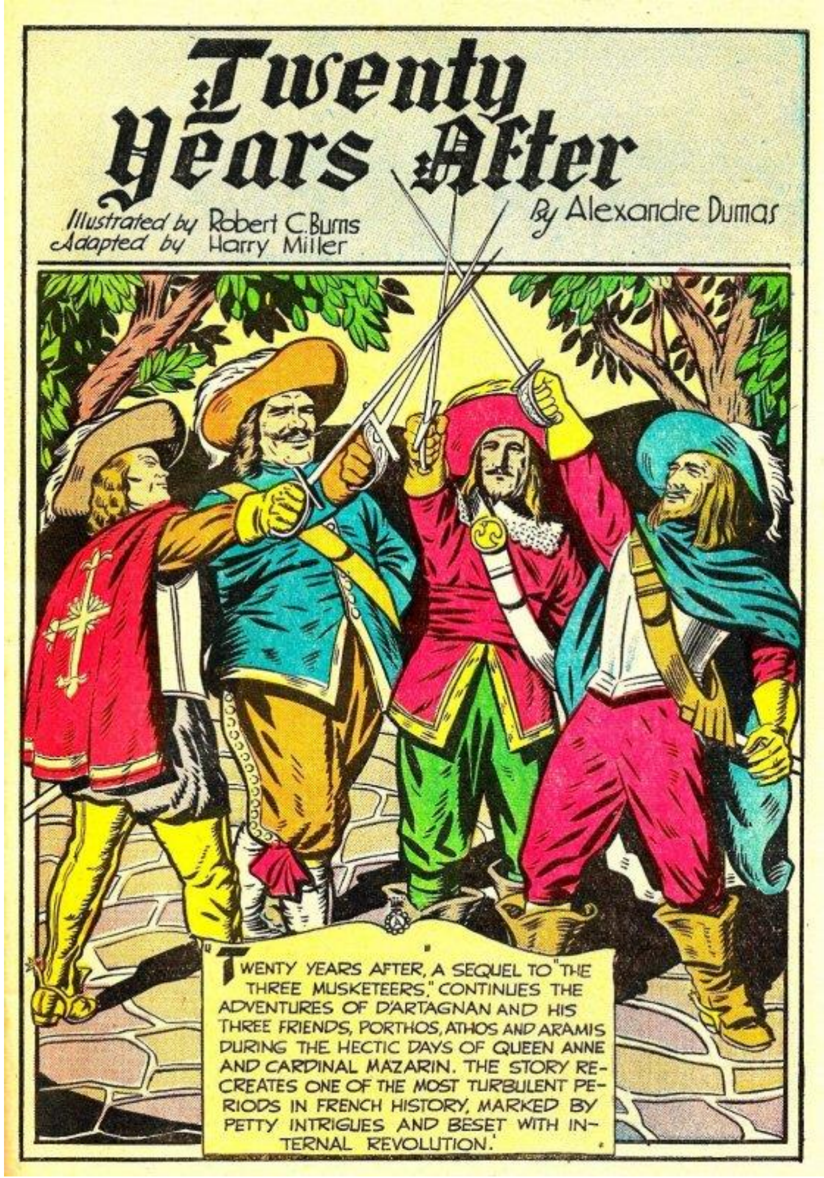




Later on, it was found that CF_3Br was harmful for the ozone layer, and was banned from production and use.

→It was replaced by C_2H_2F4, a gas largely used in refrigerating systems

→RPCs gas mixtures are nowdays almost completely made out of $C_2H_2F_4$.



Twenty years later (not by Alexandre Dumas) → …

The problem of GreenHouse Gases

- Not only CO_2 contributes to the greehouse effect. There are other gases (and families of gases) that contribute: H_20, CH_4, CFC, HCFC, NO_x, SO_x

To compare various gases, the Global Warming Potential was introduced:

- it is a measure of how much energy the emission of 1 ton of a gas will absorb over a given period of time, relative to the emission of 1 ton of carbon dioxide (CO_2).
- The larger the GWP, the more that a given gas warms the Earth compared to CO_2 over that time period.
- The time period usually used for GWPs is 100 years(*).

(*) Some gases, once arrived in the high atmosphere, dissociate, producing by-products that may or may NOT be greenhouse gases: therefore the lifetime a a gas species MUST be considered when computing its GWP

GWP for various gases

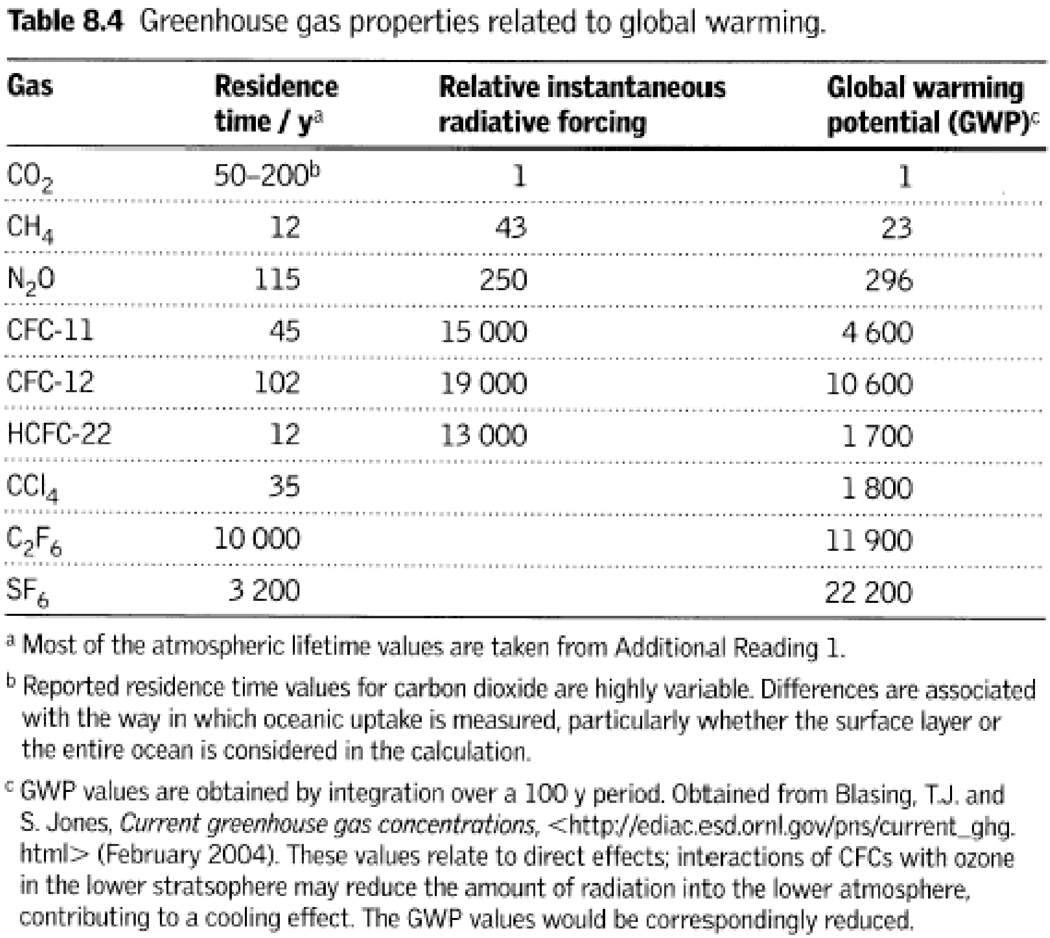


Table 8.4 Greenhouse gas properties related to global warming.

Gas	Residence time / y[a]	Relative instantaneous radiative forcing	Global warming potential (GWP)[c]
CO_2	50–200[b]	1	1
CH_4	12	43	23
N_2O	115	250	296
CFC-11	45	15 000	4 600
CFC-12	102	19 000	10 600
HCFC-22	12	13 000	1 700
CCl_4	35		1 800
C_2F_6	10 000		11 900
SF_6	3 200		22 200

[a] Most of the atmospheric lifetime values are taken from Additional Reading 1.

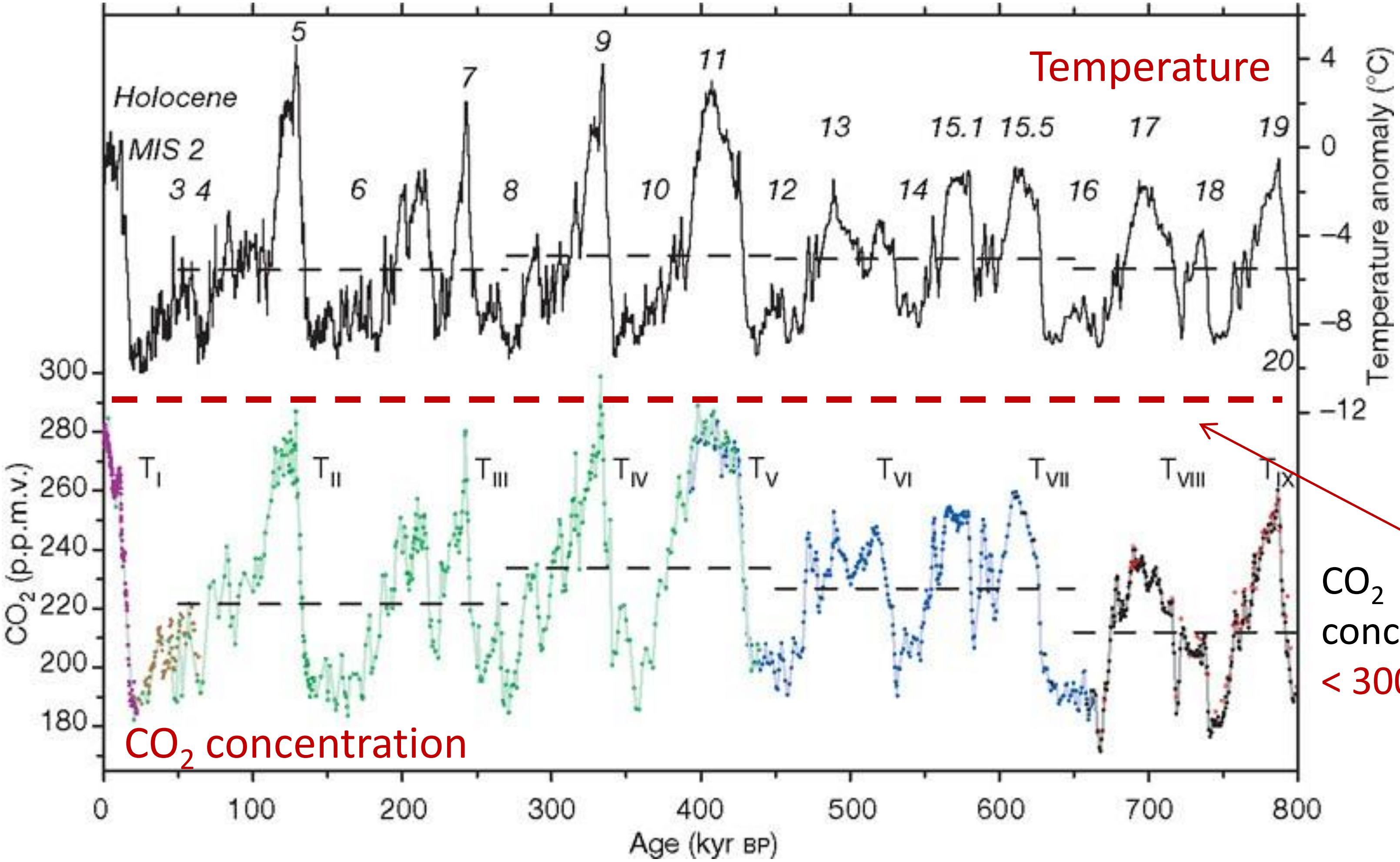
[b] Reported residence time values for carbon dioxide are highly variable. Differences are associated with the way in which oceanic uptake is measured, particularly whether the surface layer or the entire ocean is considered in the calculation.

[c] GWP values are obtained by integration over a 100 y period. Obtained from Blasing, T.J. and S. Jones, *Current greenhouse gas concentrations*, <http://ediac.esd.ornl.gov/pns/current_ghg.html> (February 2004). These values relate to direct effects; interactions of CFCs with ozone in the lower stratsophere may reduce the amount of radiation into the lower atmosphere, contributing to a cooling effect. The GWP values would be correspondingly reduced.

The Relative instantaneous radiative forcing, is independent of the lifetime of the gas in the atmoshpere.

The GWP take also into account the lifetime of the gas in the atmoshpere.

Temperature and CO_2 concentration: last 800.000 years



Temperature

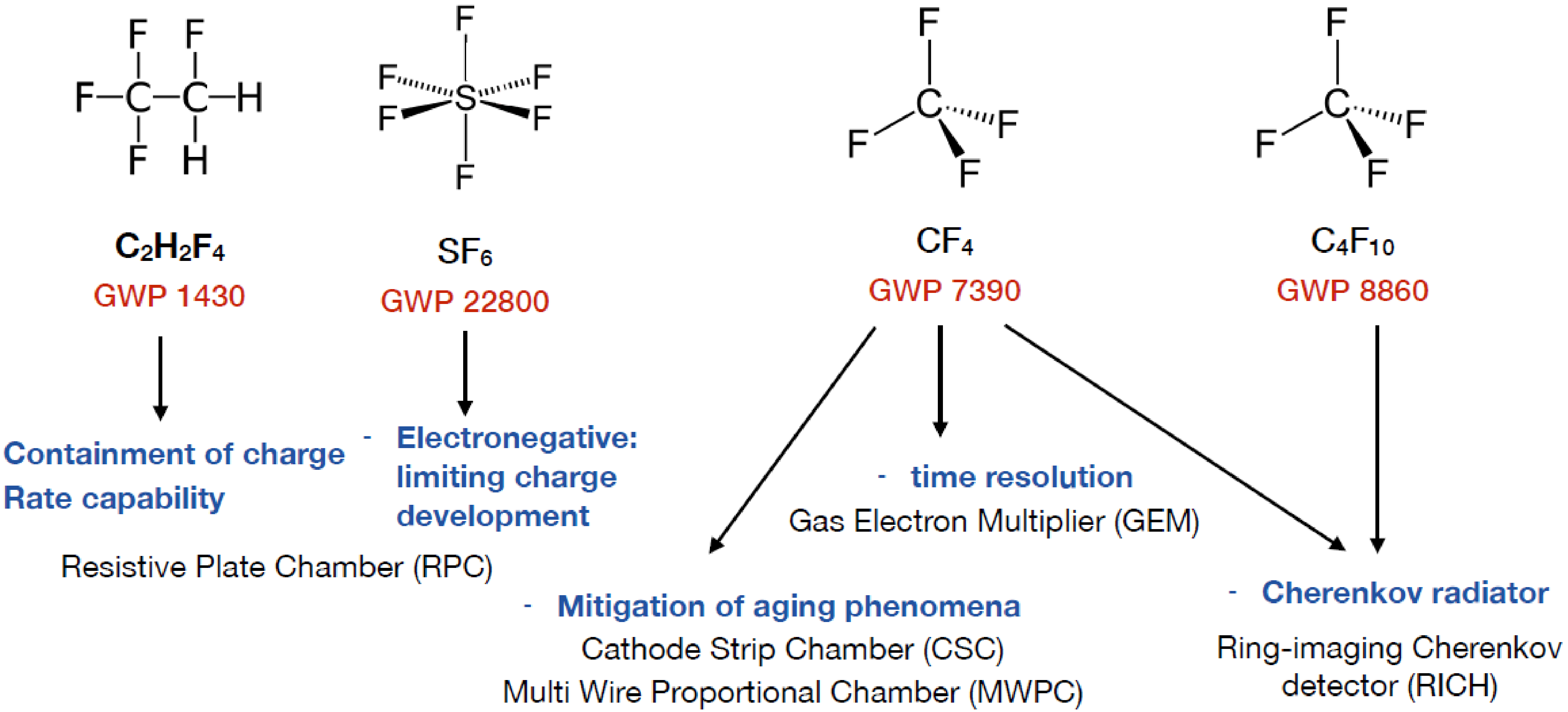
CO_2 concentration

CO_2 concentration
< 300 ppm

Lüthi, D., *et al.* High-resolution carbon dioxide concentration record 650,000–800,000 years before present. *Nature* **453**, 379–382 (2008). https://doi.org/10.1038/nature06949

Use of Greenhous gases in HEP

GHGs are used in several gaseous particle detectors due to their characteristics suitable for optimal detector performance AND long term operation



$C_2H_2F_4$
GWP 1430

Containment of charge
Rate capability

Resistive Plate Chamber (RPC)

SF_6
GWP 22800

- **Electronegative: limiting charge development**

CF_4
GWP 7390

- **time resolution**

Gas Electron Multiplier (GEM)

- **Mitigation of aging phenomena**

Cathode Strip Chamber (CSC)
Multi Wire Proportional Chamber (MWPC)

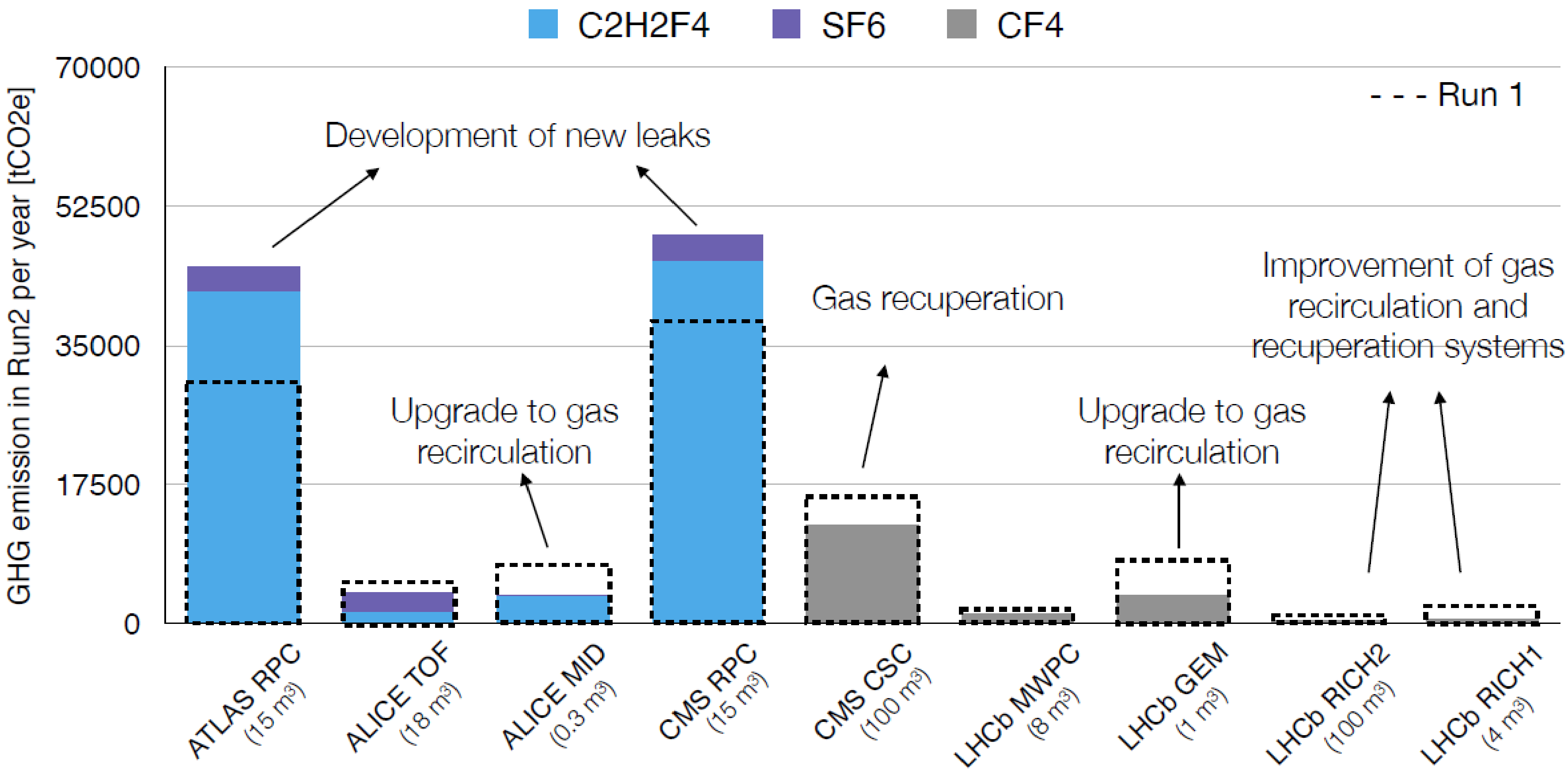
C_4F_{10}
GWP 8860

- **Cherenkov radiator**

Ring-imaging Cherenkov detector (RICH)

- 30 years ago we had to get rid of Ozone Depletion gases
- There was not the feeling about GHGs → many detectors were designed from the beginning to be used with GHGs

The use of GHGs at the LHC experiments



- 40% GHG emissions from Run 1 to Run 2 excluding from the calculations ATLAS and CMS RPC systems

- ATLAS and CMS RPC systems: +35% increase of GHG emissions due to development of new leaks

Replacement of GHGs with eco-friendly gases

We need to replace:

- ✓ $C_2H_2F_4$ = R134a = TFE mainly used in RPCs
- ✓ SF_6 mainly used in RPCs
- ✓ CF_4 used in CSCs, GEMs, RICH, etc.

It's not a problem concerning just the RPC community

with more ecological gases, namely with a much lower Global Warming Potential.

Difficult problem: gases are the core of gas-filled detectors.

We also need:

- to get the same performance
- not to change the electronics and HV (for existing systems)

➢ HEP experiments, present and future, last several (dozens) of year
→ A good performance must be mantained for an adequate period of time
→ Aging tests are needed as well.

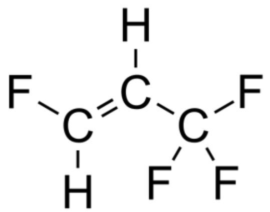
Of course we can also re-circulate the gases used, after purifying them, and reduce leaks

The importance of collaborative effort

- All high energy experiments (ALICE, ATLAS, CMS, LHCb, etc.) and the CERN gas group (CERN EP-DT) started, already several years ago, an intense R&D program to find suitable gas mixtures.

- Practically all research trendlines concentrate around the idea of replacing:
  $C_2H_2F_4$ (GWP=1430)→ $C_3H_2F_4$ze (GWP=4)
  + CO_2 (GWP=1) or He

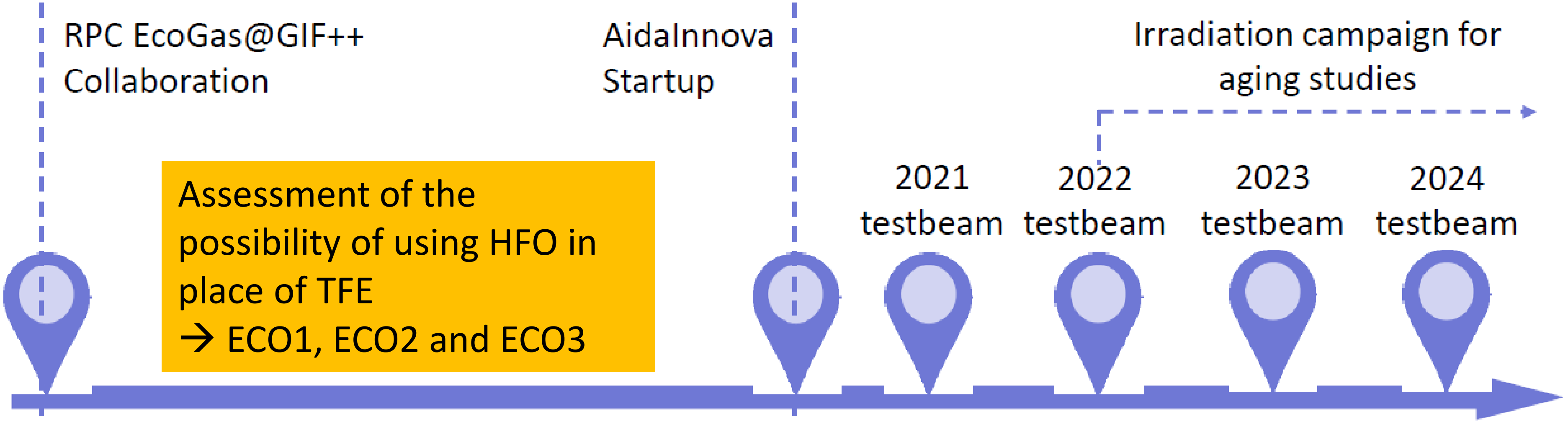
EEE



- ✓ $C_3H_2F_4$ (here indicated as HFO for short) is the molecule most similar to TFE but with low GWP
- ✓ CO_2 (or He) are essentially added to reduce the operating voltage.

The RPC EcoGas@GIF++ is a Collaboration **transversal to ALICE, ATLAS, CERN EP-DT, CMS, and LHCb** willing to put together expertise and resources in order to test potential candidates of eco-friendly gas mixtures with different detectors and electronics.

The RPC ECOGas@GIF++ timeline



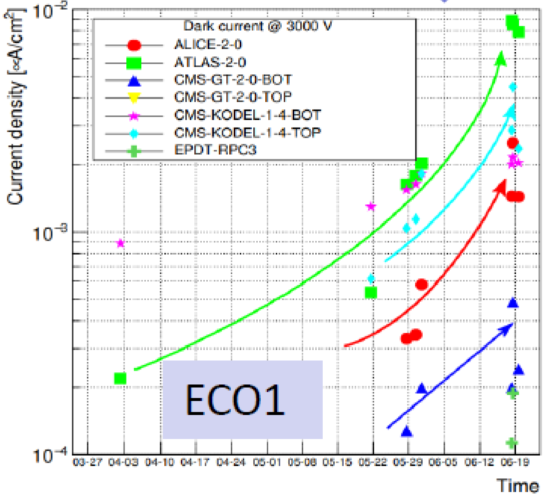
RPC EcoGas@GIF++
Collaboration

AidaInnova
Startup

Assessment of the possibility of using HFO in place of TFE
→ ECO1, ECO2 and ECO3

2018

Studies in different laboratories
Setup of the system at GIF++ and first HFO/CO_2 based gas mix. under irradiation



ECO1 discarded due to high current increase after ~20 mC/cm^2 integrated charge

2021

Irradiation campaign for aging studies

2021 testbeam

2022 testbeam

2023 testbeam

2024 testbeam

Performance baseline

Performance comparison

Std, ECO2 and **ECO3** mixtures tested under irradiation. First results in papers *[1][2][3]*.

[1] "High-rate tests on resistive plate chambers operated with eco-friendly gas mixtures", 2024 Eur. Phys. J. C.

[2] "Performance of thin-RPC detectors for high rate applications with eco-friendly gas mixtures", 2024 Eur. Phys. J. C.

[3] "Preliminary results on the long term operation of RPCs with eco-friendly gas mixtures under irradiation at the CERN Gamma Irradiation Facility" – submitted to EPJplus.

Experimental set-up @GIF++

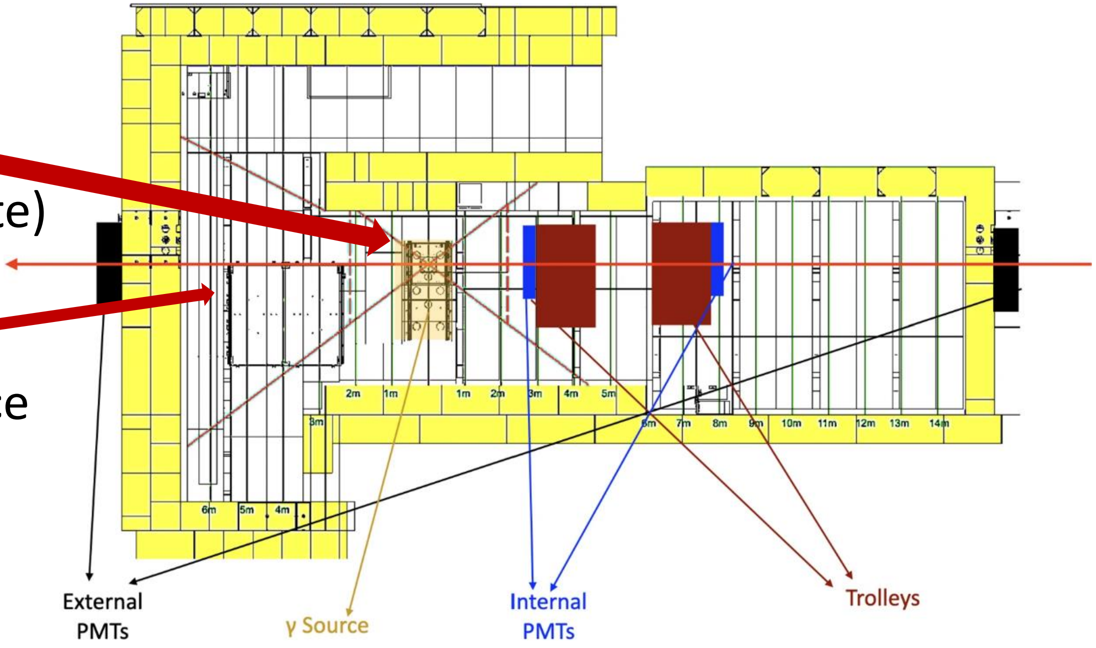
- Three gas mixtures identified, with various concentrations of HFO and CO_2.

ECO1: 45% HFO / 50% CO_2 / 4% iC_4H_{10} / 1% SF_6
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

GWP reduced by 1/3 w.r.t. the std mixture!

Attention focussed on ECO2 and ECO3 because of the good stability and performance demonstrated in home-labs tests.

Aging tests performed at GIF++
- 12.5 TBq 137Cs source
  - ✓ to generate background (high rate)
  - ✓ to accelerate aging processes
- 100 GeV muon beam
  - ✓ to measure detector performance



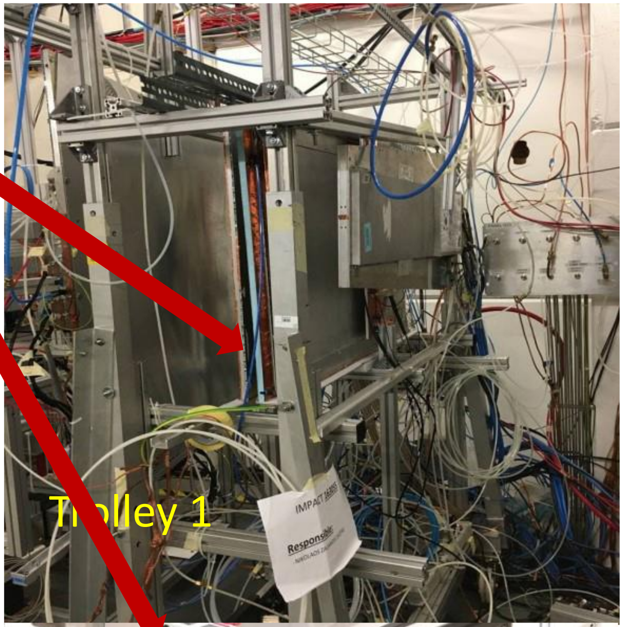
Detectors @ GIF++

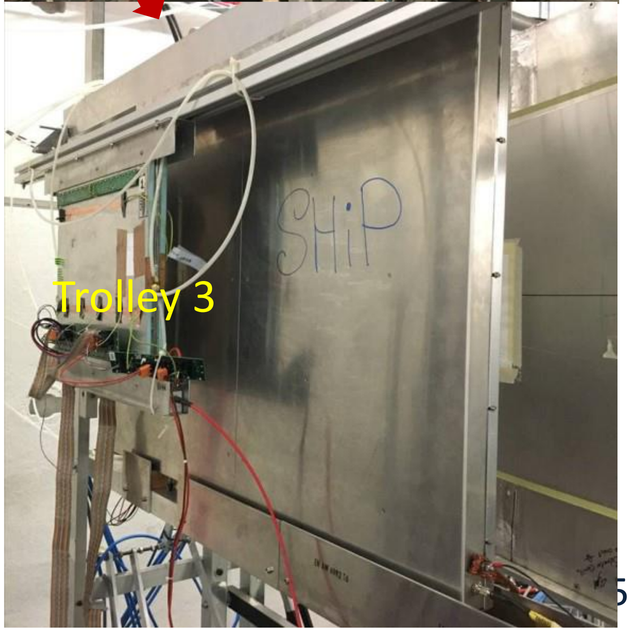
- Various detectors, mounted on two trolleys, equipped with various electronics.

→ Help in disentagling common observed effect from effects specific of ONE detector

RPC	Gap thickness	Electronics
ALICE	2mm	FEERIC + TDC
ATLAS	2mm	Digitizer
CMS	2mm – double gap	CMS FEB + TDC
CMS upgrade	1.4mm – double gap	CMS FEB + TDC
EP-DT	2mm	Digitizer
LHCb/SHiP	1.6mm	FEERIC + TDC

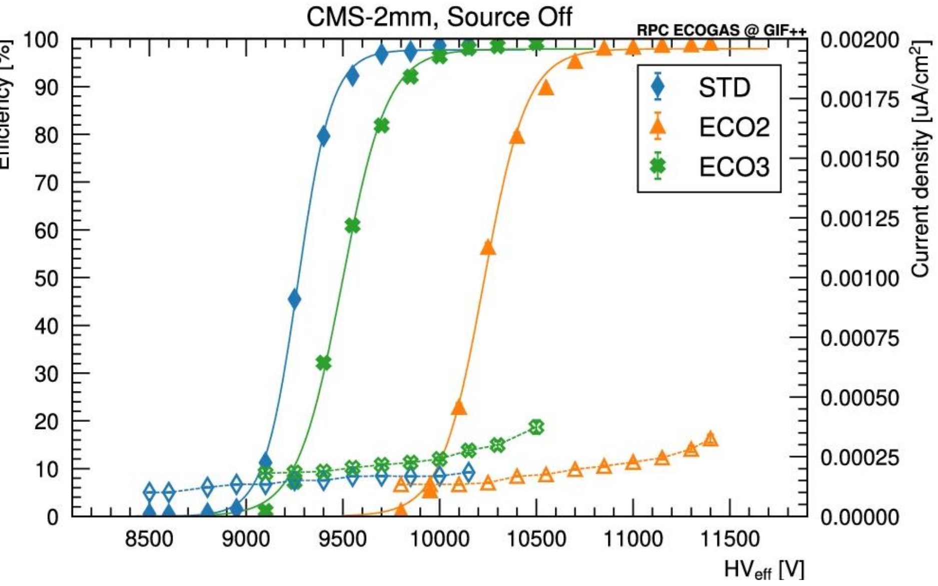
- The results presented here refer particularly to the detectors equipped with TDC

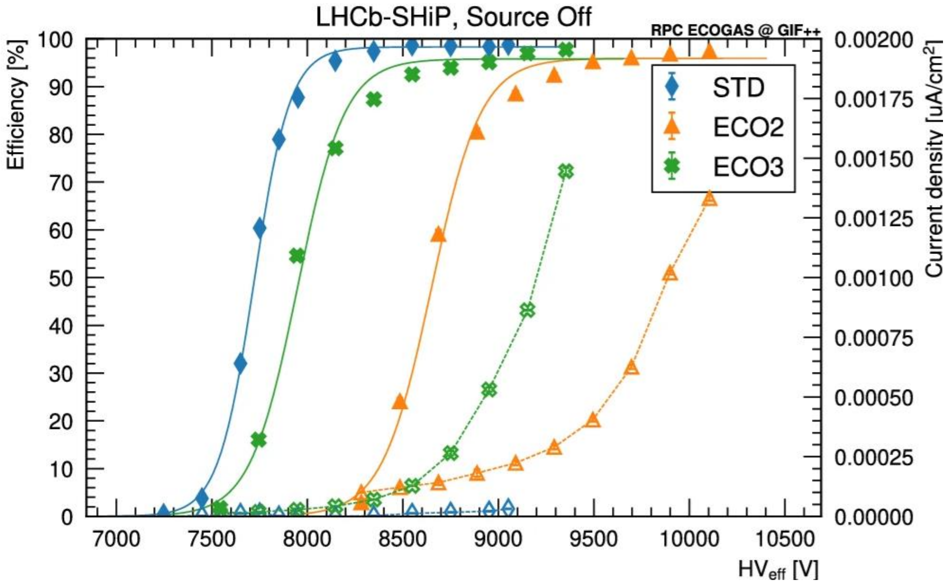




Determination of baseline perfomance

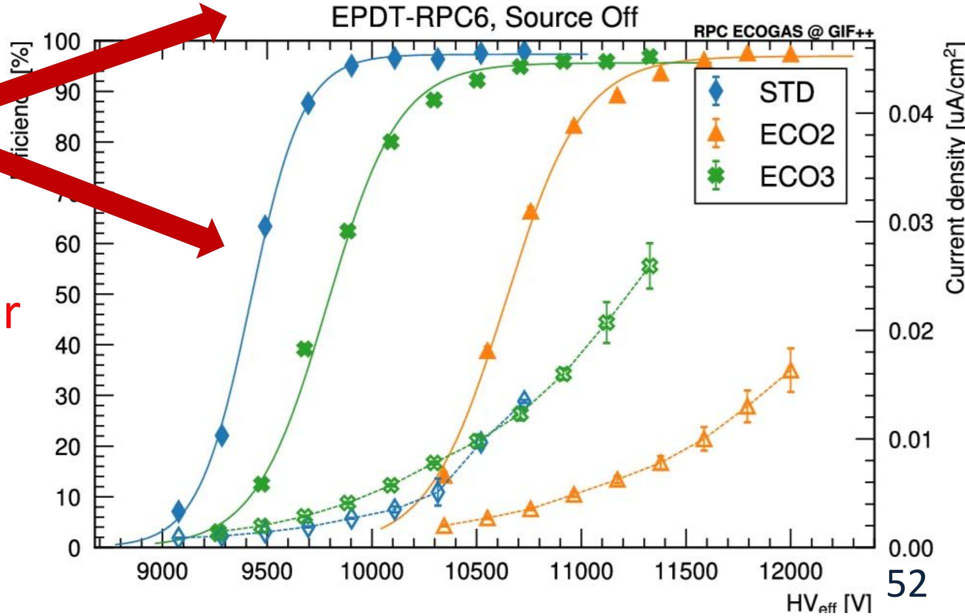
Source OFF, 2021 data


CMS-2mm, Source Off


LHCb-SHiP, Source Off

Fit of efficiency curves with a sigmoid:

$$\mathcal{E}(HV_{\text{eff}}) = \frac{\mathcal{E}_{\max}}{1 + e^{-\beta(HV_{\text{eff}} - HV_{50})}}$$


EPDT-RPC6, Source Off

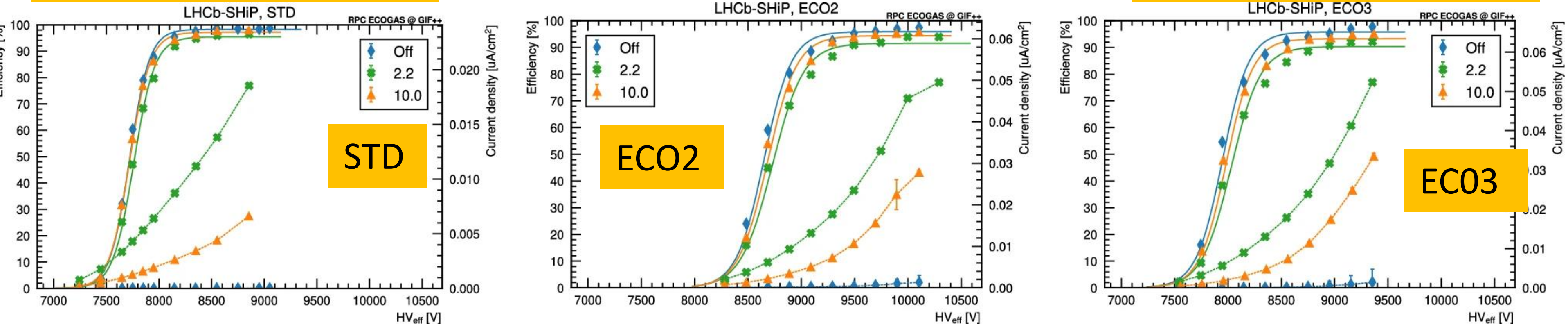
- ➢ Efficiency at plateau basically similar for STD, ECO2 and ECO3 > 95%
- ➢ Efficiency curves shifted at higher voltages depending on fraction of HFO

→ WP(ECO2) > WP(ECO3) > WP(STD)

Efficiency and counting rate with irradiation

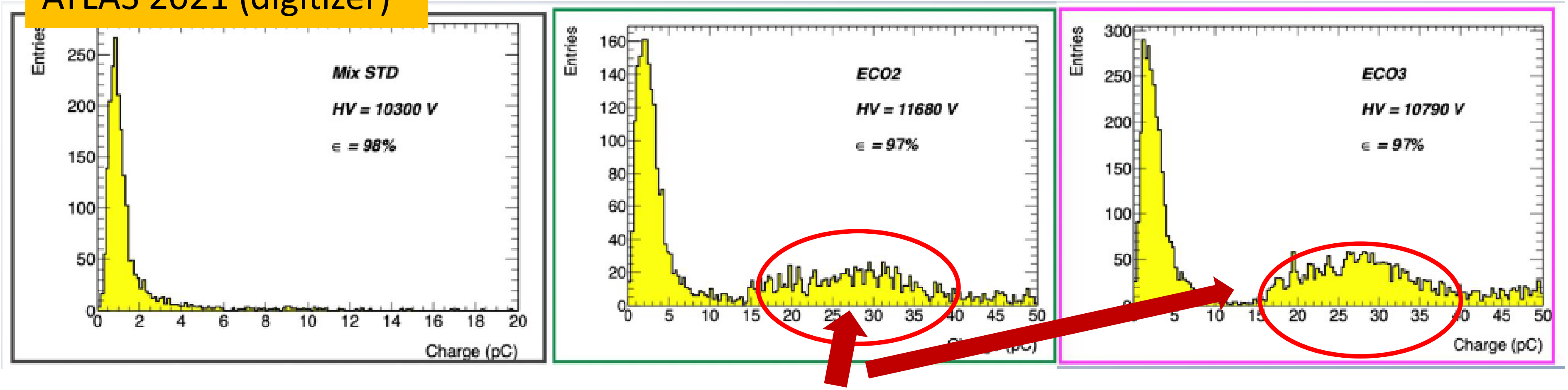
Source ON, 2021 data

LHCb/SHiP (SG 1.6 mm)



- ➢ Efficiency > 90% at the highest rate for all detectors

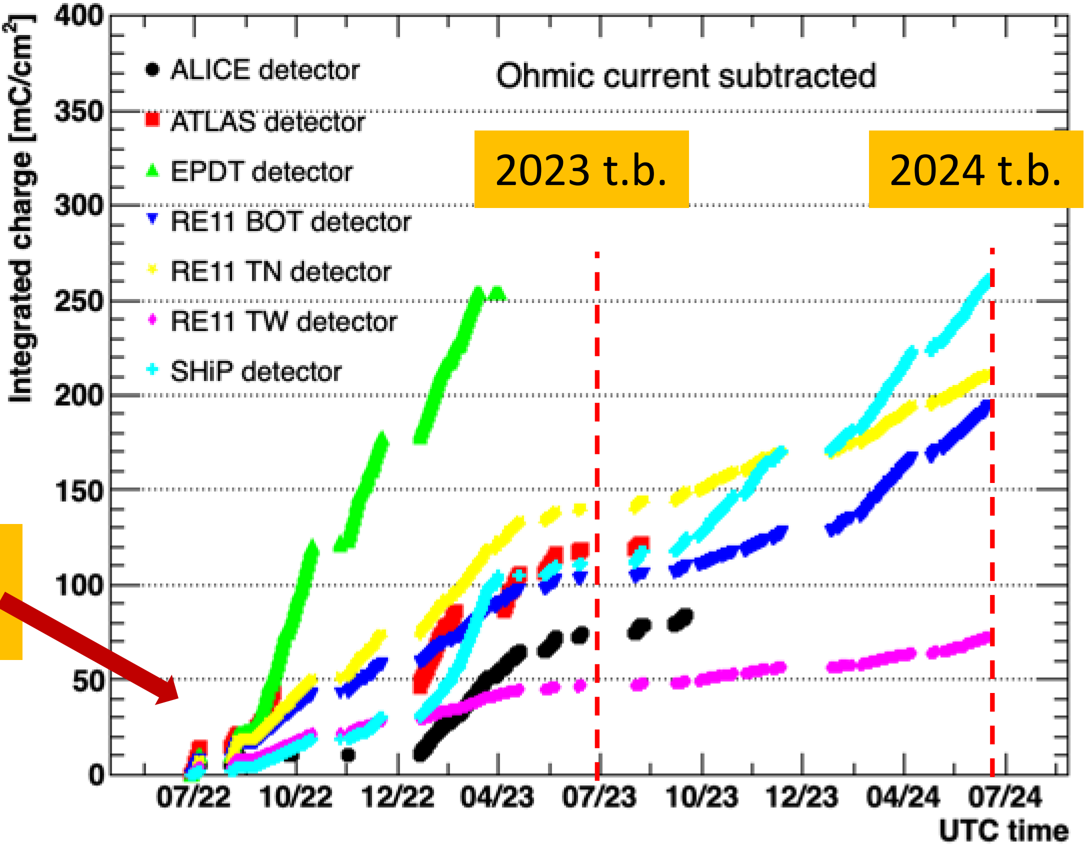
ATLAS 2021 (digitizer)



With ECO2 and ECO3 the presence of larger charge events (not streamers) observed

Aging and causes for aging in RPCs

- Generally, the charge integrated along a certain elapsed time is considered the most important factor for aging in RPCs;
- The targets of integrated charge are different for various experiment: for instance, for ALICE is ≈ 100 mC/cm², for CMS is ≈ 1 C/cm² CMS, including a safety factor of 3.



2023 t.b.

2024 t.b.

Start of the irradiation

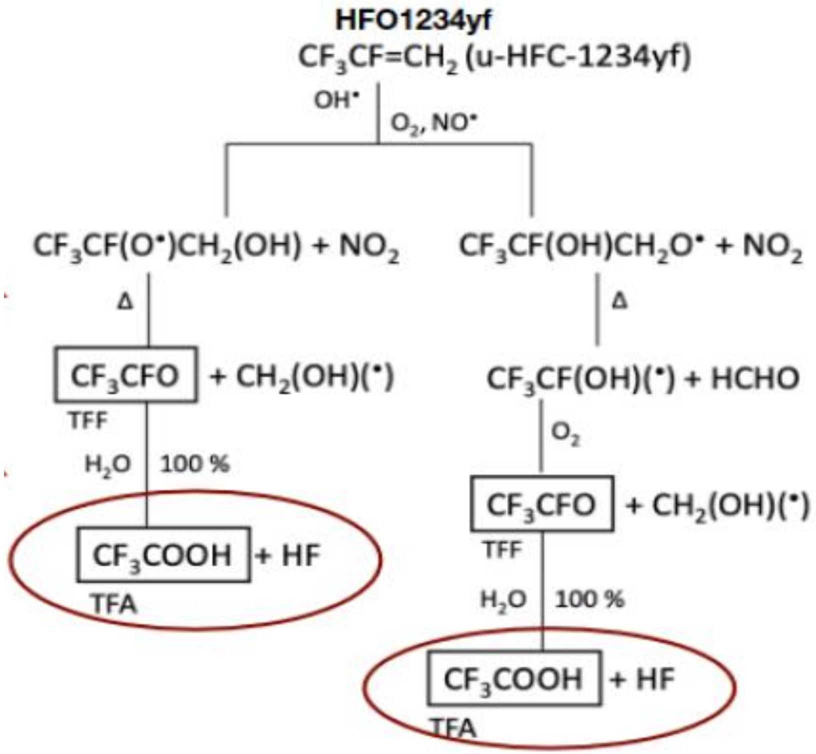
Further considerations about aging

Caveat: The importance of the integrated charge derives from the fact that production of HF was measured to be proportional to the integrated charge

→ Direct damage of the detector

➢ However, HFO typically dissociates producing TriFluoroAcetic acid

How TFA causes aging in RPCs and affects their performance on the long term is still to be investigated.

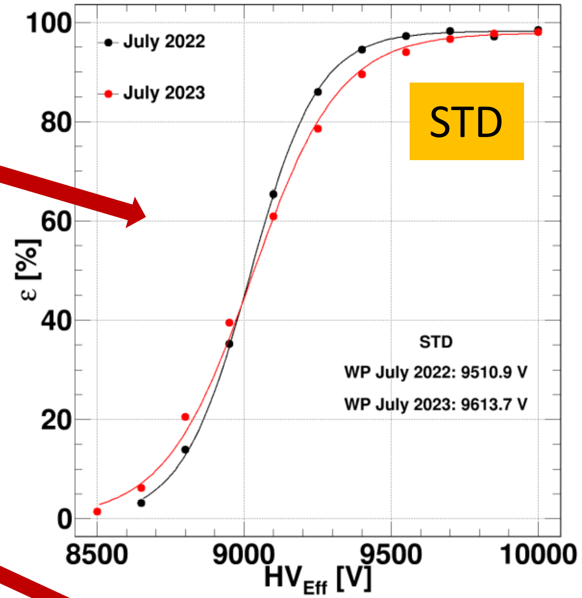


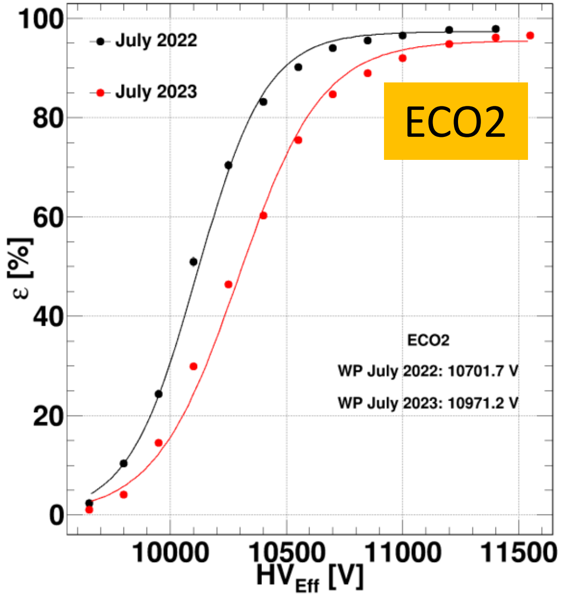
Aging is also caused by irradiation itself
→ Chemical modifications in the HPL electrodes

Aging is also caused by time itself
→ e.g. changed in HPL resistivity because of drying up.

Efficiency before and after irradiation campaign

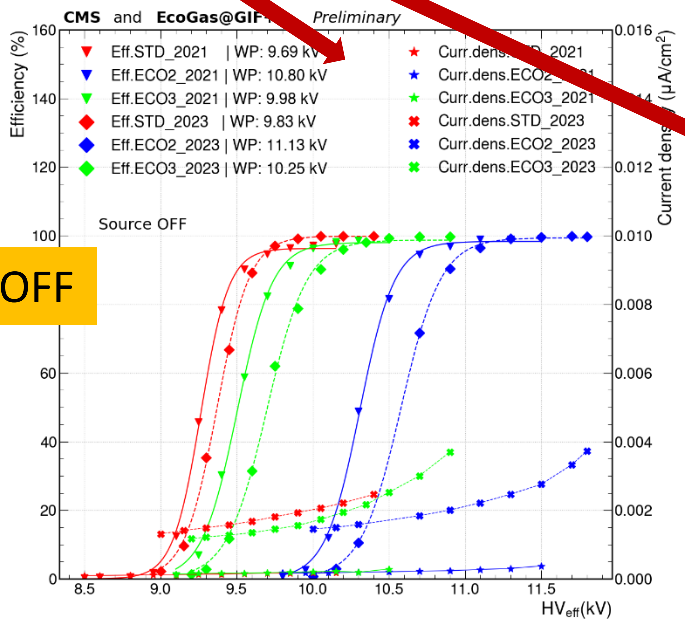
ALICE, comparison
2022 – 2023
SOURCE OFF

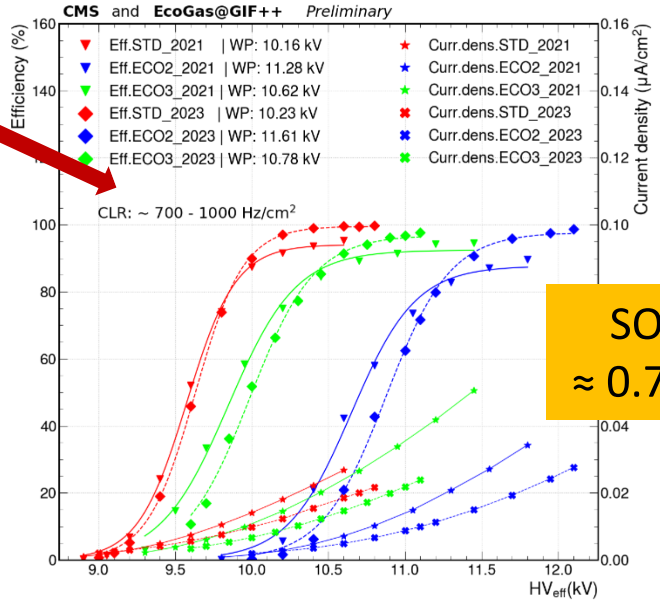




CMS (double gap),
comparison 2021 – 2023

SOURCE OFF

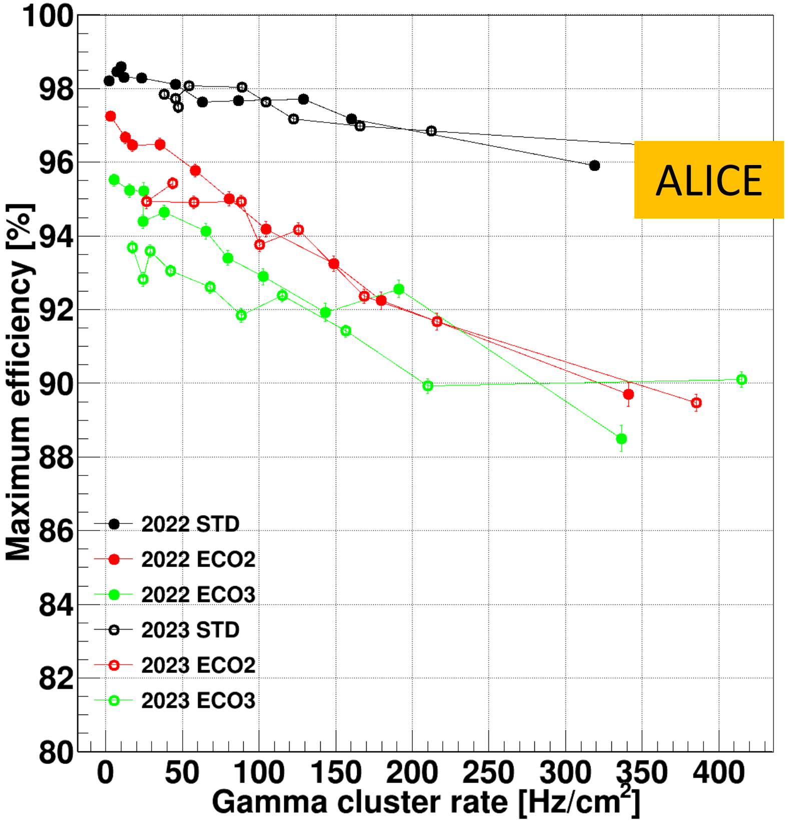


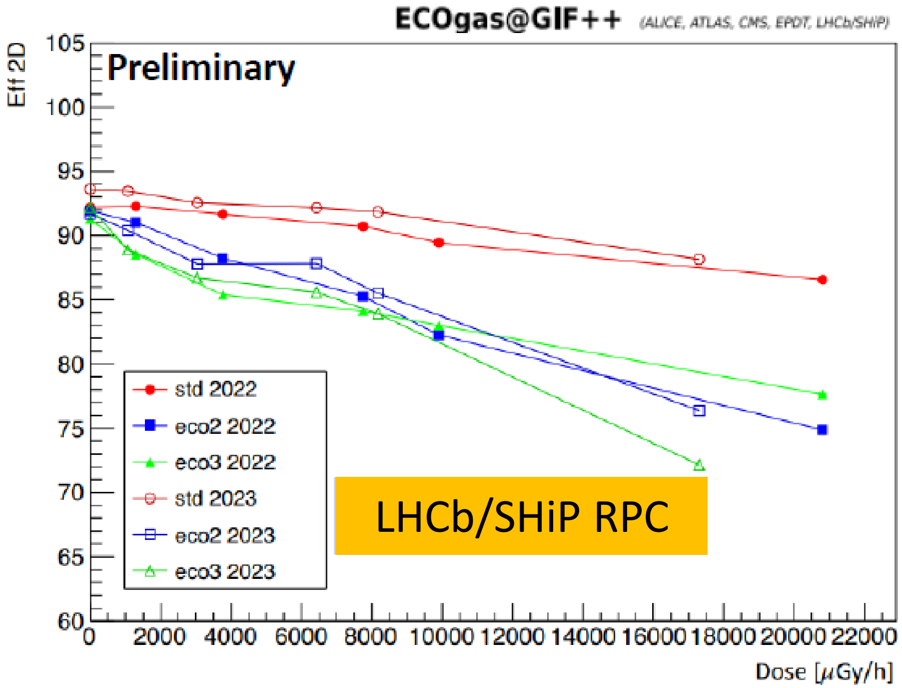


SOURCE ON
≈ 0.7-1 kHz/cm^2

Plateau efficiency before and after irradiation

Comparison 2022-2023 data





- ➢ The usual decrease of plateau efficiency with rate (or dose) is observed.
- ➢ Nevertheless, there seems NOT to be any efficiency degradation in the time lapse 2022-23

The other piece of the puzzle

- The replacement of TFE is just **part of the problem**; in ECO2 and ECO3 the residual GWP is **almost ALL due to the presence of SF6.**

- Gas mixture replacement is generally done at constant number of gas volumes

  → CO2e is the parameter to consider when evaluating the reduction of the impact on greenhouse effects

Mixture	GWP (100 y)	CO2e (g/l)
Standard	1485	6824
ECO2	475	1522
ECO3	527	1529

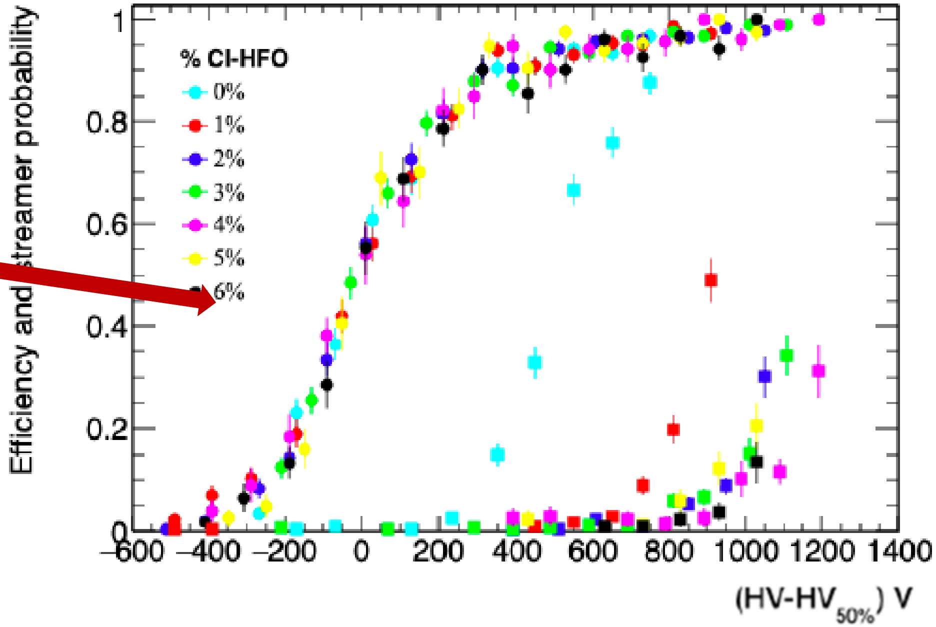
- With ECO2 and ECO3 achieved a reduction of 4 times the CO2e wrt. STD
- The residual CO2e is ALL due to SF6

→ Need to find replacement for SF_6, with low GWP and CO_2e, which could reduce the fraction of large charge events when in combination with HFO.

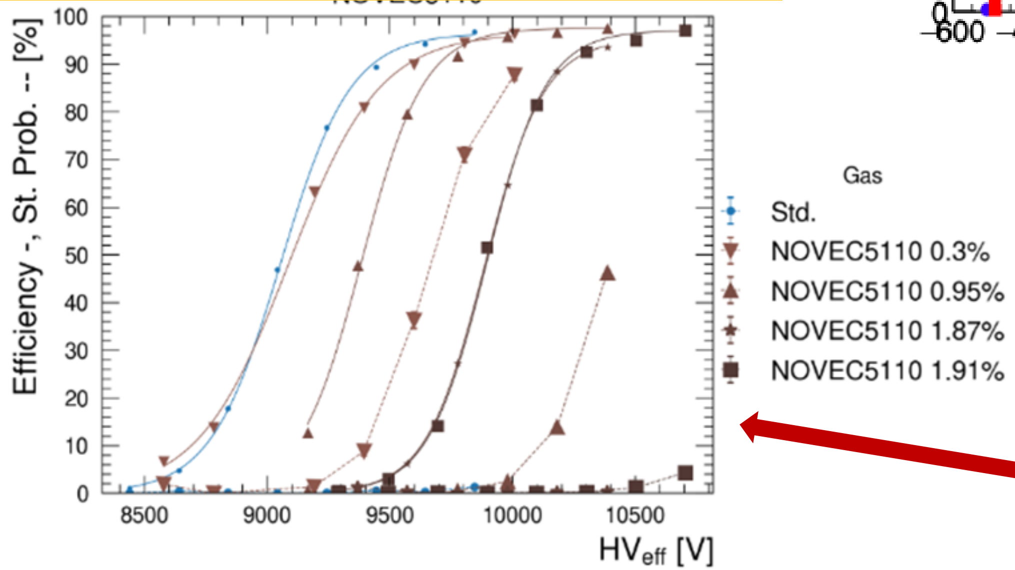
Looking for replacements for SF_6

One proposed solution is
Chloro-trifluoropopene $C_3H_2ClF_3$
(HFO1233zd)
Avalanche/streamer separation
(“useful” plateau) **larger than 400V**

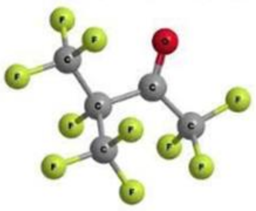
G.Proto et al, 2022 *JINST* **17** P05005



B. Mandelli, Possible alternatives to SF6 for Resistive Plate Chambers RPC2022



NOVEC 5110
- GWP < 1
- Atm. lifetime = 15 days
- Application in industry



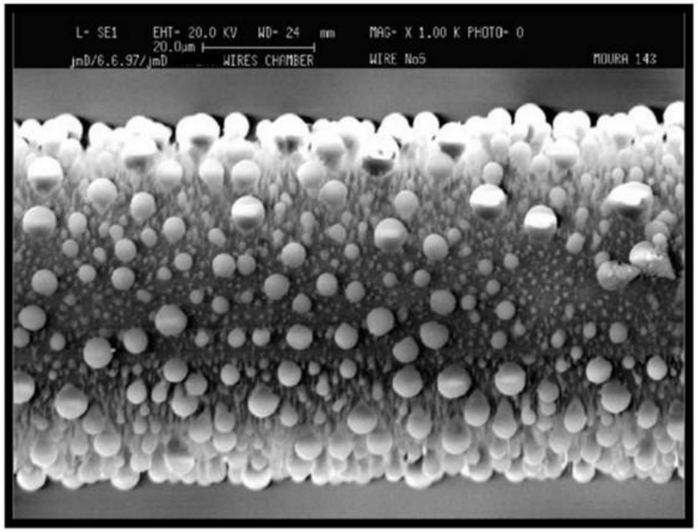
What about CF_4?

Used in CSCs and GEMs

For instance, gas mixture used in CSCs of CMS:

- **40% Ar + 50% CO2 + 10% CF4**

- The main purpose of CF4 in the gas mixture – protection against anode wire aging : Si + 4 F → SiF4
  (also breaking C-chains in polymer formation)

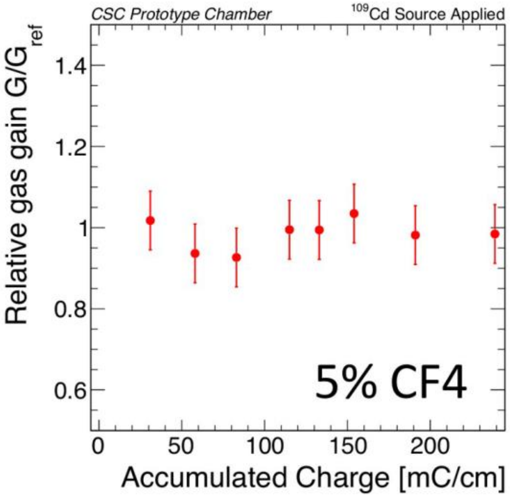


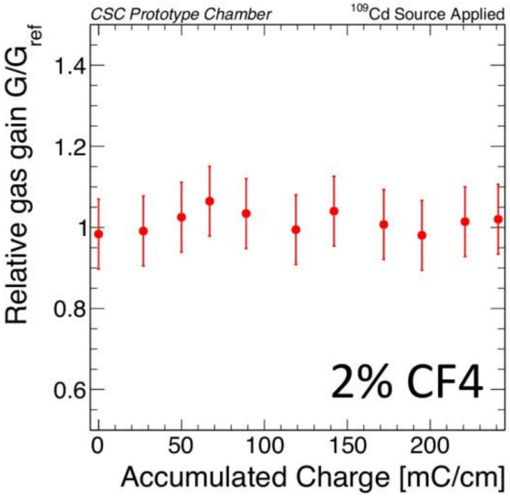
Used in GEMs basically to increase drift velocity → better time resolution. Anyhow without CF_4 time resolutions still within requirements,

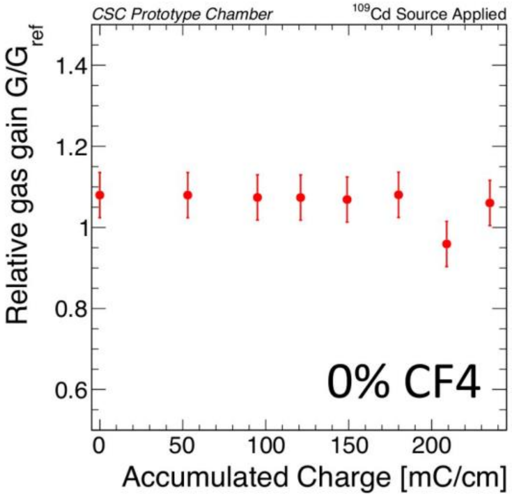
Main ideas:

- ✓ Reduce (or eliminate) CF_4
- ✓ HFO to replace CF_4, but this implies an increased HV → more studies needed

Reduction of CF_4 in CSC

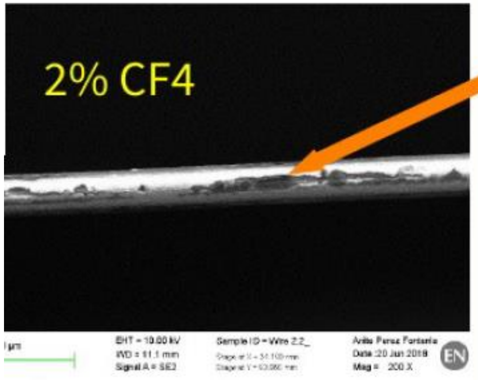


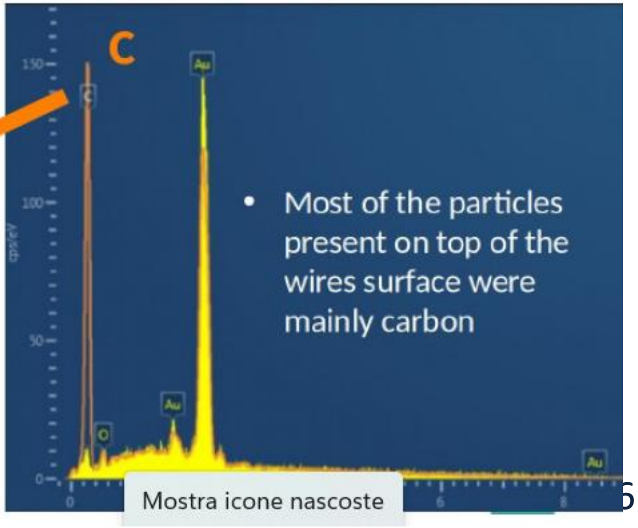




No significant degradation seen, <u>in terms of performance</u>, in all longevity tests

- However cathode modifications were seen in all cases.
- Anode surface depositions are seen with 0 and 2% CF4 even with naked eye.

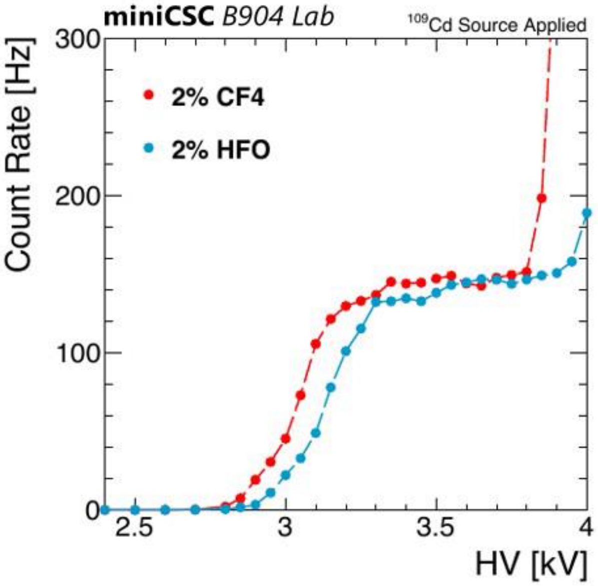


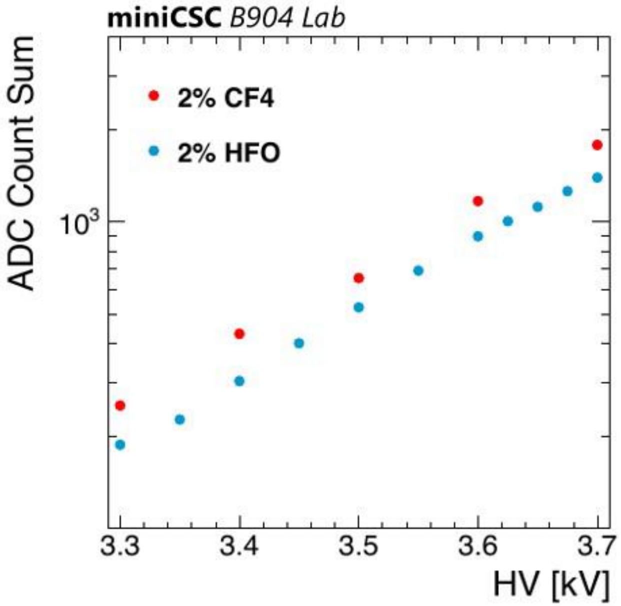


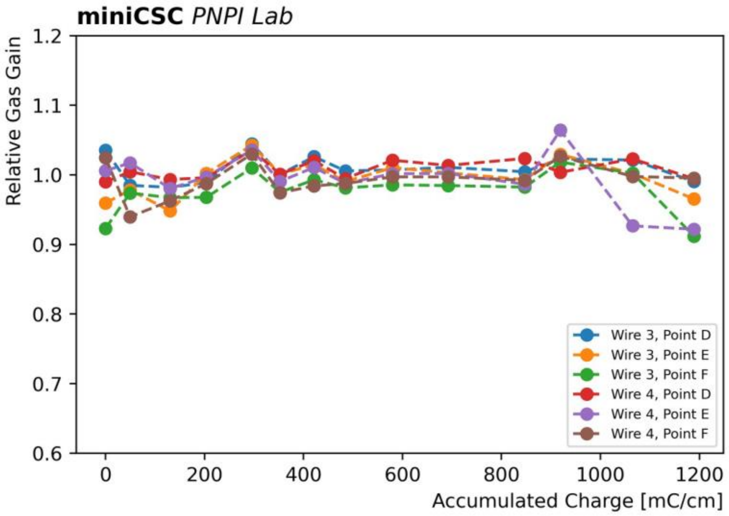
- Most of the particles present on top of the wires surface were mainly carbon

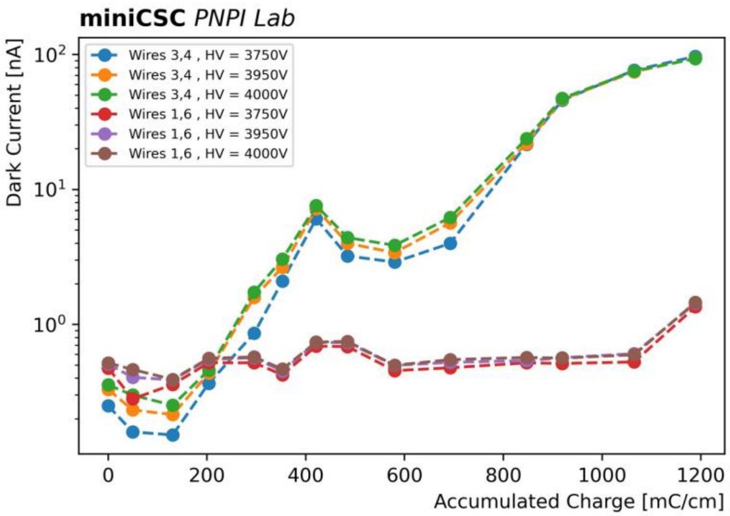
Replacement of CF_4 with HFO1234ze

Just 1000V increase in the operating voltage, good efficiency, reasonable plateau length









No gain reduction up to 1 C/cm, but significant increase in the dark current in first irradiation tests

Conclusions

- There are no conclusions!
  - Activities are in full steam, there is a LOT to do!
  - Join in, be part of the party!
- Interpretation of the effects observed not trivial
- Collaborative effors of paramount importance at this stage.

The gaseous detector community is on the eve of its ecological transition