

Green use of fluorocarbons in Cherenkov detectors and silicon tracker cooling systems: challenges and opportunities.

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"Green" fluorocarbons for Cherenkov detectors & silicon tracker cooling systems: challenges and opportunities

- Saturated fluorocarbons (PFCs, SFCs: C_nF_(2n+2)) chosen for optical properties as Cherenkov radiators: C₄F₁₀ & CF₄ in COMPASS & LHCb RICH1&2.
- Non-conductivity, non-flammability, non-toxicity & radiation resistance

➔ ideal coolants

- C₆F₁₄ used in all LHC experiments,
- C₃F₈ evaporatively cools ATLAS silicon tracker, TOTEM;
- These fluids however have high GWPs (GWP₂₀ > 5000*CO₂);
- <u>Fluoro-ketones (C_nF_{2n}O)</u>, while not industrialised over a wide C_nF_{2n}O range, can offer similar performance at very low, or 0 GWP;
- Radiation tolerance & thermal performance of 3M NOVEC 649 (lin-C₆F₁₂O) sufficiently promising to be considered by CERN to replace C₆F₁₄ for cooling, but acid production and materials compatibility issues to resolve;

Molecular shapes and GWP (1)





From: 2021-22 CERN environmental Report report https://doi.org/10.25325/CERN-Environment-2023-003

	GROUP	GASES	tCO ₂ e 2021	tCO ₂ e 2022
	Perfluorocarbons PFC (PFCs) (particle d	Loss around 10- etection and de	12 tonnes tector cooli	68 989 ng)
	Hydrochlorofluorocarbons (HFCs)	HFC-23 (CHF ₃) HFC-32 (CH ₂ F ₂) HFC-134a ($C_2H_2F_4$) HFC-404a HFC-407c HFC-410a HFC-507	36 557	86 211
Environment	Other F-gases	SF ₆ , WP 23000	16 838	18 355
Environment Report 2021–2022	Hydrofluoroolefins (HFO)/HFCs	R-449 R1234 ze NOVEC 649 GWP	86	199
CERNY		CO2	13 771	10 419
	Total Scope 1		123 174	184 173

Adding some conclusion here, in case I run out of time.

"Green" fluorocarbons for Cherenkov detectors & silicon tracker cooling systems: challenges & opportunities

- Saturated fluorocarbons (SFCs: $C_n F_{(2n+2)}$) chosen for optical properties as Cherenkov radiators: $C_4 F_{10}$ & CF_4 used in COMPASS & LHCb RICH1&2.
- Non-conductivity, non-flammability & radiation resistance

 ideal coolants
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 - C₃F₈ evaporatively cools ATLAS silicon tracker, TOTEM.
- These fluids however have high GWPs (GWP₂₀ > 5000*CO₂).
- While not industrialised over a wide C_nF_{2n}O range fluoro-ketones can offer similar performance at very low, or 0 GWP;
- Radiation tolerance & thermal performance of 3M NOVEC 649 (lin-C₆F₁₂O) sufficiently promising to be considered by CERN to replace C₆F₁₄ for cooling;
- <u>Cherenkov detectors</u>: subject to optical testing, *lin-C₄F₈O could replace C₄F₁₀* while *lin-C₄F₈O or lin-C₅F₁₀O blended with N₂ and monitored in real time by sound* velocity gas analysis - could replace CF₄
- <u>SI tracker COOLING</u>: Lighter molecules (e.g. C₂F₄O, with similar thermodynamics to C₂F₆) would allow lower temperature, 0GWP operation than evaporative CO₂ in Si trackers operated at high luminosity: low order C_nF_{2n}O toxicity a problem...

What gives NOVEC 649/1230 (lin C₆F₁₂O: FK-5-1-12) its low GWP?



A: Structure!: a *double-bonded oxygen atom on a peripheral spur* of the molecule

This fluoro-ketone configuration is: $CF_3CF_2C(O)CF(CF_3)_2$

What gives NOVEC 649/1230 (lin C₆F₁₂O: FK-5-1-12) its low GWP?

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Scission by UV photons of λ ~300 nm in upper atmosphere (low P, high UV): fragments do not re-associate* into C_nF_(2n+2) SFCs

*The Environmental Impact of CFC Replacements HFCs and HCFCs <u>T. WALLINGTON</u> et al *Environ.Sci.Technol.*1994(28)7 320A https://doi.org/10.1021/es00056a714

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The Cherenkov detector aspect

Ideally... replace SFCs with sameorder GWP₀ FKs with good UV transparency, refractivity...



Sustainability session: ICHEP2024 Prague 18-24 July 2024

Refractive index & GWP 'load' in a Cherenkov Gas Radiator (vessel only, not including tankage & piping)

C₄F₁₀ (COMPASS: 100m³, LHCb RICH 1: 4m³): *GWP*₂₀ = 4880, CF₄ (LHCb RICH 2: 100m³): *GWP*₂₀ = 6870

Cherenkov radiator vessel volume $V(m^3)$ filled with a blend of gases of densities ρ_i (kgm⁻³),

fractional concentrations $w_i \&$ individual GWP_i (tonnes $CO_2 eq.)$ has a GWP environmental "load" (& release potential) L given by:

$$L = \frac{V}{1000} \sum_{i} (wi \cdot \rho_i \cdot GWPi) \quad \text{(tonnes CO}_2 \text{ eq.)}$$

Corresponding radiator gas mixture refractivity given by :

$$(n-1)_{rad} = \sum_i (wi.(n-1)_i)$$

Can blend small concentration ω_x of (heavier) SFC or Lin-FK vapour of high refractivity $(n-1)_x$ with ω_y of light transparent gas (N₂, Ar...) to replicate the target refractivity $(n-1)_x$ of a lighter SFC at high concentration – for lower GWP load.

$$\omega_{x} = \frac{(n-1)_{z(target)} - (n-1)_{y}}{(n-1)_{x} - (n-1)_{y}}$$

Use real-time speed of sound to get component molar concentrations, ω_i (SoS traditionally also called "c" by acousticians to confuse things)

$$c = \sqrt{\frac{\gamma RT}{M}} \ \gamma_m = \frac{C_{pm}}{C_{vm}} = \frac{\sum_i w_i C p_i}{\sum_i w_i C v_i} \quad M = \sum_i w_i M_i \qquad c = \sqrt{\frac{\sum_i w_i C p_i}{\sum_i w_i C v_i} RT}{\sum_i w_i M_i}$$

More details (ATLAS system): https://www.mdpi.com/2410-390X/5/1/6

Then use standard refractivity formula to get from calculated $\omega_{1,2}$

$$(n-1)_{rad} = \sum_i (wi.(n-1)_i)$$

to refractive index of radiator gas blend in real-time, then standard relativistic expressions to get from n to Cherenkov γ thresholds for different particle species + β = 1 angle

Real-time measurement of speed of sound c takes us via the relative concentrations of the components to the speed of light β and beyond!! in the radiator gas

Eyes on the prize...(focus here on LHCb RICH2 (n-1)...) (488.10⁻⁶)_{CF4} = (~1750.10⁻⁶)_{C5F100}*0.12+(300.10⁻⁶)_{N2}*0.88

Cherenkov threshold: *lin*-C₅F₁₀O (eg: 3M NOVEC 5110)/N₂ & (heritage) C₅F₁₂/N₂ blend

and GWP load comparison with LHCb RICH2 (CF_4 : 100m³)



Measured Sound Velocity (m/s)

Radiator volume GWP load: 100 m³ gas radiator with different mw gases blended with nitrogen to replace CF₄

(note: blends with Ar very similar: need refractivity data for linear fluoro-ketones, probably similar to SFCs) blends with CO_2 not consdered: high CO_2 triple point temp (-56 C) \rightarrow thermodynamic recirculation difficulty)



Even bigger GWP load reduction through C₄F₁₀ index matching (eg here: COMPASS RICH)

Table 1: GWP loads of various SECs and NOVEC 5110 blended with N₂ ((n-1) = 310.10⁻⁶) to match refractivity of CF₄ and C₄F₁₀ assumed radiator volume: 100 m³

Base fluid	Base fluid density (1bar,25°C) kgm ⁻³	Base fluid GWP (20-yr)	Component (n-1) (*10 ⁶) (@ nm)	% Blend with N ₂ to match (n-1) CF ₄	GWP load (t.CO ₂)	% Blend with N₂ to match (n-1) C₄F ₁₀	GWP load (t.CO ₂)
CF ₄ LHCb RICH2 (100m ³)	3.56 [18]	4880 [16]	488 (180-310 nm) [19]	100	1737	not applicable	n/a
CF ₂ O	-			~100	0	n/a	n/a
C ₂ F ₆	5.63 [18]	8210 [16]	793 (180-310 nm) [19]	38.1	1762	not applicable	n/a
Lin-C ₂ F ₄ O	-			-		n/a	n/a
C ₃ F ₈	7.75 [18]	6640 [16]	1180 (250 nm) [16]	21.4	1099	not applicable	n/a
Lin-C ₃ F ₆ O	-			-		n/a	n/a
C ₄ F ₁₀ LHCb RICH1 (4m ³): COMPASS (100m ³)	9.97 [18]	6870 [16]	1450 (250 nm) [16]	16.3	1119	100	6849
Lin-C ₄ F ₈ O (Non-cyclic C ₄ F ₈ O)*	9.5 (est.)	Probably < 1 (NOVEC 5110 Analogy)	1380 @ 400nm (based on 3M PFG-3480 c- C_4F_8O [7]: linear C_4F_8O not yet measured but assumed similar)	18.4	0.18	112.7^* (>100% would imply necessity of operating C_4F_8O at slight overpressure)	1.07
C ₅ F ₁₂	11.63 [18] (BP 30 °C at 1 bar)	6350 [16]	1750 (180-310nm)[19] (40 °C, undiluted)	13.0	957	79.3	5857
NOVEC 5110 C₅F ₁₀ O	10.7 [13] (BP 27 °C at 1 bar)	<1 [13]	Not yet measured: probably around 1650 by analogy with C_4F_{10} and C_4F_8O ratio	13.9	0.149	85.2	0.91

G. Hallewell: DRD4 WG 2 Low GWP FC radiator gases: June 19 2024





Some molecular shapes to study (or eliminate) for UVT, GWP, radiation stability & materials compatibility: example: C_4F_8O isomers,

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²¹ UV transparency, refractivity, cmhemical stability & compatibility studies needed for different C_nF_{2n}O molecular shapes.

RICH detector community very competent & equipped for detailed UVT & (n-1)







Synquest Inc. FL, USA

On the turning away...

The uncertain ECHA (European Chemicals Agency) route to fluorocarbon (PFC, PFAS...) prohibition (Other people here will talk about this...Path paved with impracticalities?)

[51] ECHA/NR/23/04;
https://echa.europa.eu/-/echa-publishes-pfas-restriction-proposal]
[52] ECHA Candidate List of substances of very high concern for Authorisation;
https://echa.europa.eu/candidate-list-table
[53] Annex to the Annex XV restriction report proposal for restriction: Per- & polyfluoroalkyl substances (PFASs); ECHA; 22/03/2023
https://echa.europa.eu/documents/10162/d2f7fce1-b089-c4fd-1101-2601f53a07d1
[54] Per- and polyfluoroalkyl substances (PFAS); ECHA
https://echa.europa.eu/hot-topics/perfluoroalkyl-chemicals-pfas

Au revoir 3M... need to clarify manufacturers' attitudes (Dehon Co. (Fr), Synquest (USA), F2 (UK), various companies in China)... on future *lin*-fluoroketone (C_nF_{2n}O) production Electronics industry is the driver: we just ride the coat-tails!

The Silicon tracker cooling aspect

See also

https://link.springer.com/article/10.1140/epjp/s13 360-023-04703-w

²⁴ Examples from cooling particle physics Si trackers (ATLAS, CMS, LHCb)

Two distinct types of detector cooling geometry at HL-LHC:

(1) Tube and block 'DNA'



ATLAS barrel SCT (present)

pathed pathed path

Thermal Figure of Merit (TFM):

Expressed in terms of temperature difference between Si modules & coolant at given power density (units: ^oC cm² W⁻¹)

TFM = $((T_{(Si module)} - T_{(Coolant)}) / (Si module power/cm²)).$

200 µm

Lower numeric value → better thermal performance. TFMs < 4 typical in microchannel devices, ~ 20+ in some tube & block geometries.

Disadvantages: long(er) heat conduction path (more interfaces) Si→ cold(er) coolant: lower Thermal figures of merit NEED COLDER COOLANT! (2) Micro-channel cooling

LHCb VELO upgrade

Advantages: shortest heat conduction path to coolant: coolant can be warmer: Disadvantages: fraglity issues: channels etched in silicon and cover plate attached

Micro-channel cooling: the LHCb VELO upgrade Recent mastery of evaporative CO₂ cooling in microchannels



CO₂ Evaportive cooling problems:

High triple point temperature of -56 °C

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CO₂ cooling already known to be inefficient at coolant temperatures below -40 °C:

limits the lowest temperature attainable in the tubes of a tube & block cooling system: CO₂ cooling now admitted to be insufficient for protection against radiation damage and protection against leakage current-generated thermal runaway over the whole HL-LHC envelope (2029-41):

ATLAS plans to change its inner pixel detector half way through...

A very complicated alternative trans-critical krypton evaporative cycle has been proposed

see: L. Contiero et al; Cold Krypton system for the Phase III Upgrade of the LHC, Forum on Tracking Detector Mechanics 2023, Tuebingen Germany May 31-June 2 2023

 27 Si tracker evaporative coolingome Thermodynamic comparisons: SFCs, CO₂, Xe

(F-K thermodynamics would be similar to same carbon order SFCs)



https://link.springer.com/article/10.1140/epjp/s13360-023-04703-w

 C_2F_4O would have been the ideal fluid: much cheaper than Xe, but toxic... blend C_2F_6 with C_3F_6O ?? (C_2F_6 / C_3F_8 zeotropic blends already demonstrated...) R. Bates et al; 2015 JINST 10 P03027

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[52] ECHA Candidate List of substances of very high concern for Authorisation;
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[53] Annex to the Annex XV restriction report proposal for restriction: Per- & polyfluoroalkyl substances (PFASs); ECHA; 22/03/2023
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Conclusion

(Also at beginning as I didn't think I'd get this far...)

Anyway... thanks for watching and listening:

(some references and back-up material follow for those interested in this subject)

Also see: https://link.springer.com/article/10.1140/epjp/s13360-023-04703-w

Conclusion

- Saturated fluorocarbons (SFCs: C_nF_(2n+2)) are chosen for their optical properties as Cherenkov radiators, with C₄F₁₀ and CF₄ used in COMPASS & LHCb RICH1&2.
- Non-conductivity, non-flammability & radiation resistance → ideal coolants with C₆F₁₄ used in all LHC experiments, while C₃F₈ evaporatively cools the ATLAS silicon tracker and TOTEM.
- These fluids however have high GWPs (GWP₂₀ > 5000*CO₂).
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- <u>SI tracker COOLING</u>: Lighter molecules (e.g. C₂F₄O, with similar thermodynamics to C₂F₆) would allow lower temperature, OGWP operation than evaporative CO₂ in Si trackers operated at high luminosity: low order C_nF_{2n}O toxicity a problem...

References and back-up material

Approaches to low-GWP fluorocarbon RICH radiator gases

<u>See</u>	e also:
(1)	https://link.springer.com/article/10.1140/epjp/s13360-023-04703-w
	(paper: EuroPhysics J. Plus, Dec 2023)
(2)	https://indico.cern.ch/event/1263731/contributions/5398511/attachments/2648319/4584649
/G	Hallewell DRD4%20Rad%20Gas%20GWP%20with%20annexes%20May%2016%202023.pdf
(2)	https://indice.com.ch/ovent/1/10202/ (DPD/ WC2 meeting 17 May 2024)

(3) <u>https://indico.cern.ch/event/1410802/</u> (DRD4 WG2 meeting 17 May 2024)
 (4) <u>https://indico.cern.ch/event/1420840/</u> (DRD4 WP_3.1 meeting 28 May 2024)

Cherenkov radiator and related general references (1 of 2) from

https://link.springer.com/article/10.1140/epjp/s13360-023-04703-w

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[3] B. Mandelli; R&D for the optimization of the use of greenhouse gases in particle detector systems; Mini workshop on gas transport parameters for present and future generation of experiments: CERN April 22, 2021 https://indico.cern.ch/event/1022051/contributions/4325947/

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[5] S. Dalla Torre et al; Long term experience with C₄F₁₀ in COMPASS RICH-1; Presentation 11th Intl. Workshop on Ring Imaging Cherenkov detectors, Edinburgh, Scotland, Sept 12-16 2022.

https://indico.cern.ch/event/1094055/contributions/4932286/attachments/2508724/4311387/RICH2022_C4F10_dallatorre.pdf

[6] 3M PFG-3480: c-octofluorotetrahydrofuran (C_4F_8O). **Note**: fluid out of production : product reference now used for a non-fluidic product. For historic product data sheet mentioning its high GWP see (for example):

http://static6.arrow.com/aropdfconversion/a7116f41dfdd5b79d2eb7b40afd687f8af23d8ef/mediawebserver(563).pdf

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[8] **Product #**2H07-2-08 **Synonyms:** 2,2,3,3,4,4,5,5-octafluorotetrahydrofuran, Perfluorotetrahydrofuran; **CAS No:** 773-14-8, **MDL No.** MFCD00465561: SynQuest Labs Inc., 13201 Rachael Boulevard, Alachua, FL 32615, USA

https://www.synquestlabs.com/Home/ProductDetail?SearchText=Octafluorotetrahydrofuran

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[11] 3M Novec® range of fluorinated fluids; <u>https://www.3m.com/3M/en_US/p/c/b/novec/?Ntt=novec</u>

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- [53] Annex to the Annex XV restriction report proposal for restriction: Per- & polyfluoroalkyl substances (PFASs);

ECHA; 22/03/2023 https://echa.europa.eu/documents/10162/d2f7fce1-b089-c4fd-1101-2601f53a07d1

[54] Per- and polyfluoroalkyl substances (PFAS); ECHA https://echa.europa.eu/hot-topics/perfluoroalkyl-chemicals-pfas

(5.7) SWOT analysis of cooling fluids HL-LHC

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Strength Non-flamma toxic, electrica non-ozone-o radiation ro GWP	IS CO₂ able, non- al insulator, depleting, esistant, P=1	Weaknesses CO ₂ High pressure circulation (60 bar) at ambient temp before cooldown to operating temp High triple point (-56°C)	Strengths C ₂ F ₆ Non-flammable, non- toxic, electrical insulator, non-ozone-depleting, radiation resistant	Weaknesses C ₂ F ₆ Very high GWP (around 6000 x CO ₂)	
			Threats C_2F_6	Opportunities C_2F_6	
Threats CO2OHigh triple point may makeExSi tracker operationExproblematic: less thermalEx		Opportunities CO ₂ Extensive R&D program at CERN Evaporative coolant of choice for ATLAS, CMS for start of HL-LHC program	Production will be phased out unless a strong motivation from semiconductor manufacture industry	Proved to decrease the operating temp of an ATLAS SCT thermal model in blend with 75% C ₃ F ₈	
irradiaton ('thermal					
runaway' phenomenon)			Strengths C _n F _{2n} O	Weaknesses	
Strengths xenon Non-flammable, non- toxic, electrical insulator, non-ozone-depleting, radiation resistant, GWP=0	S XENON able, non- al insulator,	Weaknesses xenon High pressure circulation	Non-flammable, non-toxic, electrical insulator, non- ozone-depleting, radiation resistant, GWP=0	C _n F _{2n} O	
	(50 bar) at ambient temp before cooldown to operating temp (amost transcritical) Extremely expensive	Threats $C_n F_{2n} O$ Large scale industrial production may depend on the phasing-out of SFCs, needs of semiconductor	Opportunities $C_n F_{2n} O$ Expertise in particle physics community for		
Threats xenon Very difficult future procurement (war in Ukraine) (10 ⁻⁸ atmosphe ric content.)		Opportunities xenon Could find expertise In particle physics community for fabrication of circulators: already used	manufacture industry: Material compatibility needs further study.	3M NOVEC 649 (C ₆ F ₁₂ O)	
			v dotactors & Si trackor oo	aling	
	Sustainability session: ICHEP2024 Prague 18-24 July 2024				
	Gusta				

The uncertain ECHA path to prohibition (a path paved with impracticalities?)



G. Hallewell: Green FCs for Cherenkov detectors & Si tracker cooling Sustainability session: ICHEP2024 Prague 18-24 July 2024

States, will then decide on the potential restriction.

ECHA > Information on Chemicals > Candidate List

Candidate List of substances of very high concern for Authorisation

(published in accordance with Article 59(10) of the REACH Regulation)

Notes:

- Authentic version: Only the Candidate List published on this website is deemed authentic. Companies may have immediate legal obligations following the inclusion of a substance in the Candidate List on this website including in particular Articles 7, 31 and 33 of the REACH Regulation.
- Numerical identifiers: Each candidate list entry covers both anhydrous and hydrated forms of a substance. The CAS
 number shown in an entry is typically for the anhydrous form. Hydrated forms of the substance identified by other CAS
 numbers are still within the scope of the entry.
- Other numerical identifiers: For those entries with "-" in the EC number and CAS number columns, a non-exhaustive
 inventory of EC and/or CAS Registry numbers describing substances or groups of substances considered to fall within the
 scope of the Candidate List entry is included, where practicably possible. This information can be accessed through the
 "Details" button of the selected entry.

FURTHER INFORMATION

- More information about Candidate list of Substances of Very High Concern for Authorisation
- Data on Candidate List substances in articles
- Reason for inclusion
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