

ISOTOPIC EXCHANGE INSIDE IMPREGNATED ACTIVATED CARBONS

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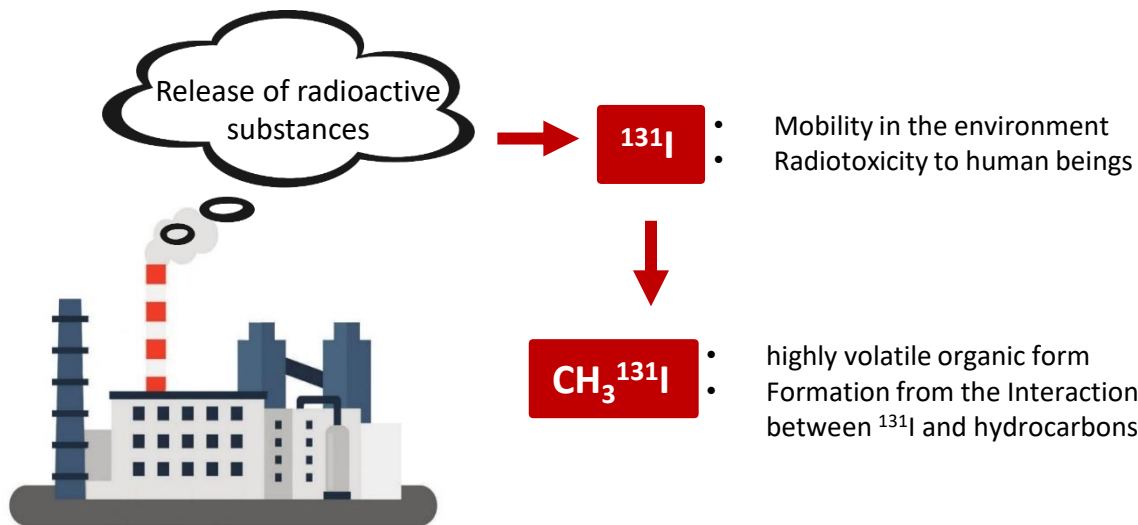
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2. Decontamination factors (DF) determination of activated carbons towards γ -labelled CH_3I
3. Breakthrough curves measurements of activated carbons towards stable and γ -labelled CH_3I
4. Conclusions and perspectives

Context



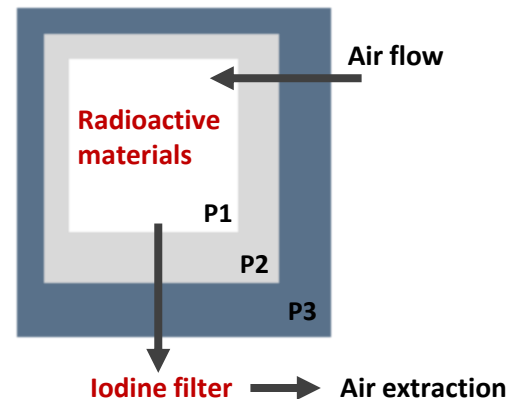
Necessity to trap $\text{CH}_3^{131}\text{I}$ through the iodine filters in the ventilation networks of nuclear facilities

- **Static containment: physical barriers**

Glove box, fume cupboard, room, building ...

- **Dynamic containment:**

Atmospheric pressure > P3 > P2 > P1



Filtration conditions for gaseous effluents in nuclear facilities

- T : < 30°C
- Relative humidity:
- Up to 40% (with upstream heater)
 - Up to ~70% (without heater)

Context

Filter of Camfil® ActiCarb
Nucléaire



Filter material for iodine trapping:
Activated carbon



Why choose activated carbons:

- Simplicity and good efficiency with impregnation
- Low maintenance and operating costs
- Reasonable price

...

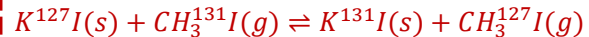
Three mechanisms for $\text{CH}_3^{131+127}\text{I}$ trapping

1. Physisorption

2. Chemisorption of $\text{C}_6\text{H}_{12}\text{N}_2$ (TEDA)

Numerous experimental studies (static, dynamic) and modeling → reaction mechanism depending on the water vapor content

3. Isotopic exchange of KI



Few studies on the mechanism of isotopic exchange in nuclear grade activated carbons

Better assess the contribution of isotopic exchange between $\text{CH}_3^{131}\text{I}$ and K^{127}I in activated carbons

Methodology

A selection of commercial activated carbons (AC):

KI impregnated AC

TEDA impregnated AC

Co-impregnated AC

Physico-chemical characterization

- Impregnation quantity
- Surface morphology
- Porous structure

1

Decontamination factors (DF) measurements of $\text{CH}_3^{131}\text{I}$

- Standardized norms*
- T = 20°C, R.H. = 40% and 90%

2

Breakthrough curves measurements

- Breakthrough curve measurements of stable and radioactive CH_3I in similar conditions

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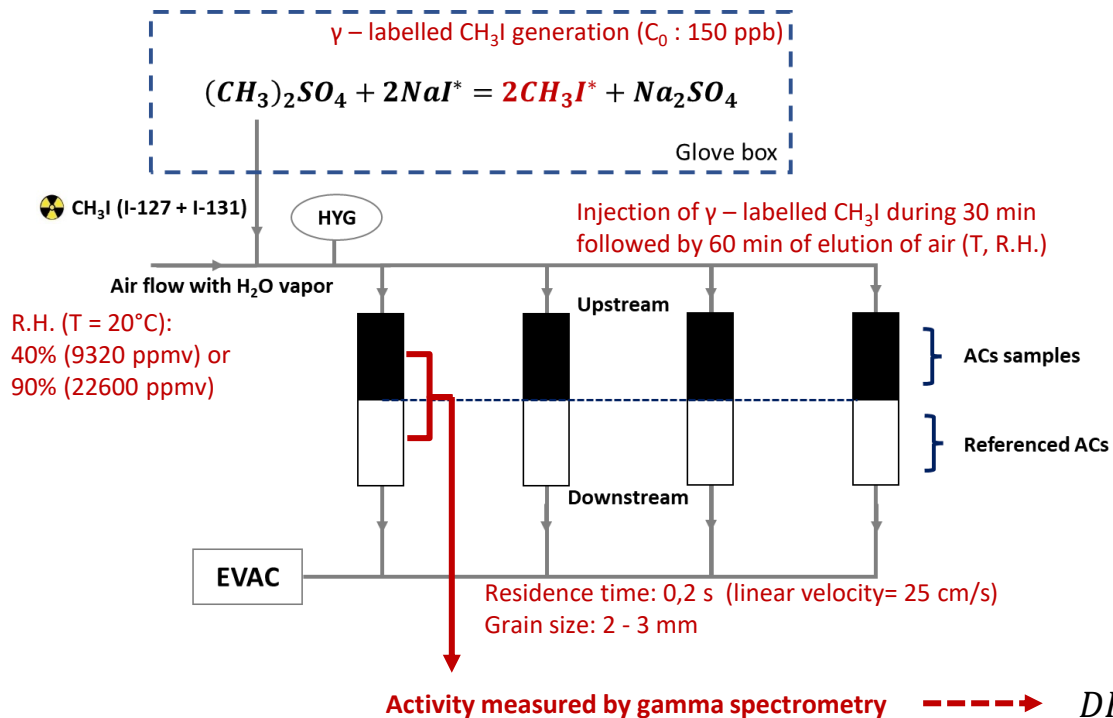
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Decontamination factors (DF) measurements of CH₃¹³¹I

Representation of PERSEE facility
(Plateforme Expérimentale de Recherches Sur l'Épuration des Effluents radioactifs)

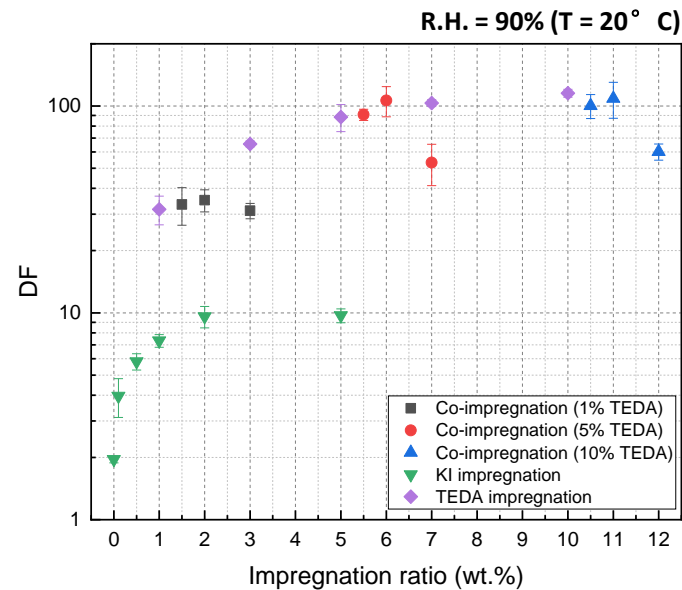
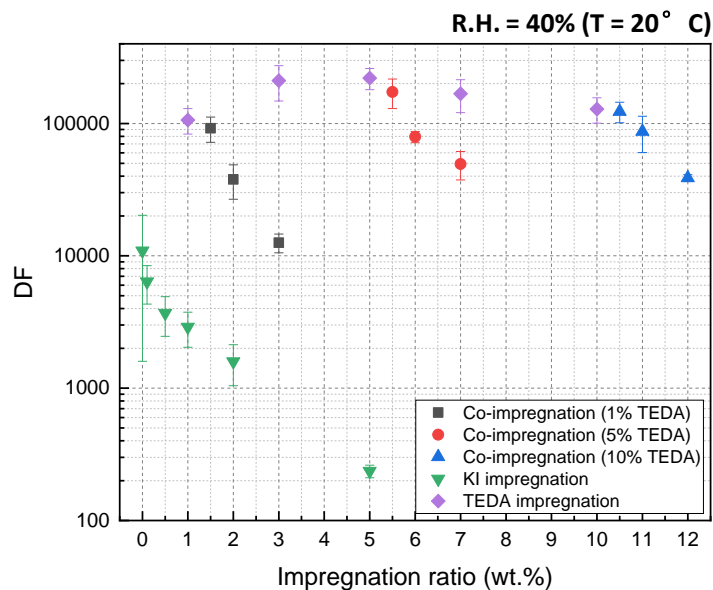


$$DF = \frac{C_{upstream}}{C_{downstream}} = \frac{A_{upstream} + A_{downstream}}{A_{downstream}}$$

Decontamination factors (DF) measurements of $\text{CH}_3^{131}\text{I}$

DF	100000	10000	1000	100	10	1
P	0.001%	0.01%	0.1%	1%	10%	100%
η	99.999%	99.99%	99.9%	99%	90%	0%

Relation between DF, P and η



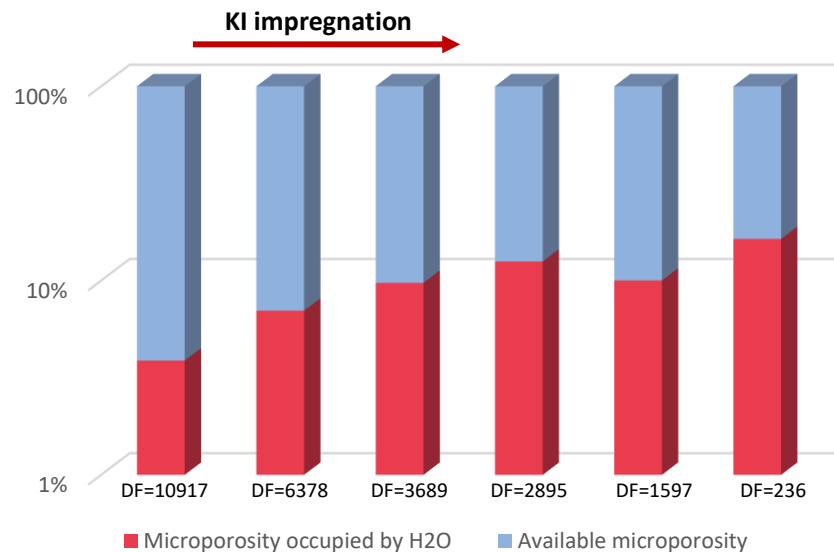
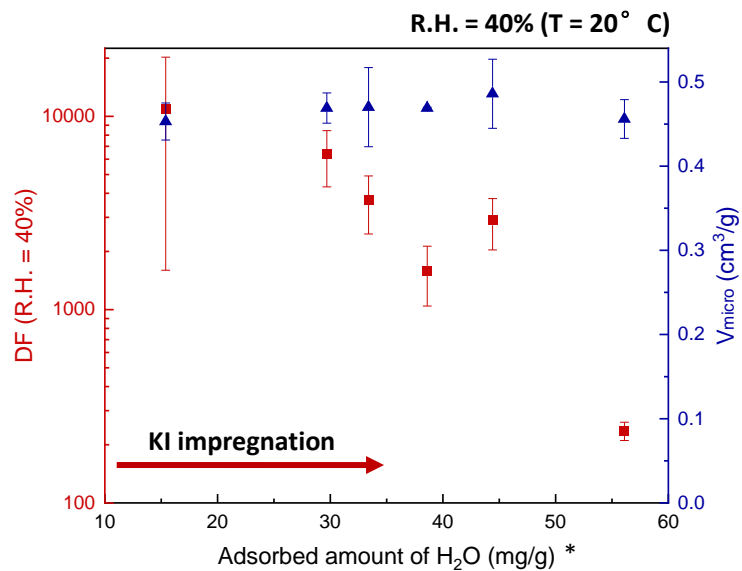
- DF : 10^4 - 10^5 : microporous characteristics of AC → retention of CH_3I by physisorption phenomena (kinetic diameter : 0.5-0.6 nm)
- TEDA impregnation (< 5wt.%) → increasing performance of CH_3I trapping → alkylation mechanism, optimum towards 5 wt.% in TEDA : compromise between chemisorption and micropore blocking phenomena
- Impregnation with KI → decrease in DF ?
- Similar DF evolutions between co-impregnated and single impregnated ACs → no interaction between KI and TEDA as demonstrated by characterization techniques

- Drastic decrease in DF : competitive adsorption between water vapor and CH_3I
- TEDA impregnation → counteract the adverse effect of high R.H. (CE~100) : protonation mechanism
- KI impregnation → gradual increase in performance (CE~10) → isotopic exchange ?
- Similar DF evolutions between co-impregnated and single impregnated ACs → no interaction between KI and TEDA as demonstrated by characterization techniques

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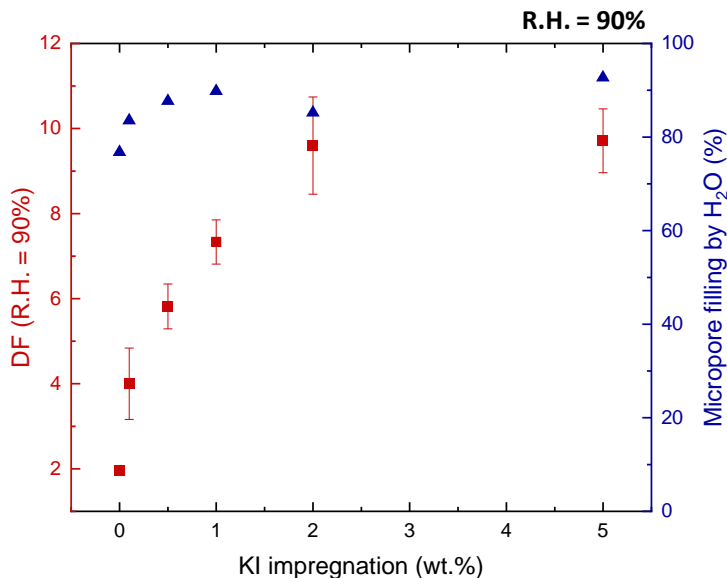


- Similar microporosity for these KI impregnated ACs due to low impregnation ratio
- Progressive KI impregnation → effect of nucleation of KI as functional groups on the AC surface → facilitate the H₂O attraction
 - increasing amount of adsorbed H₂O → decreasing available microporosity for CH₃¹³¹I trapping by physisorption
- Effect of H₂O under current conditions → determining role for physisorption
- Absence of isotopic exchange ?

Decontamination factors (DF) measurements of $\text{CH}_3^{131}\text{I}$

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Relation between DF, P and η



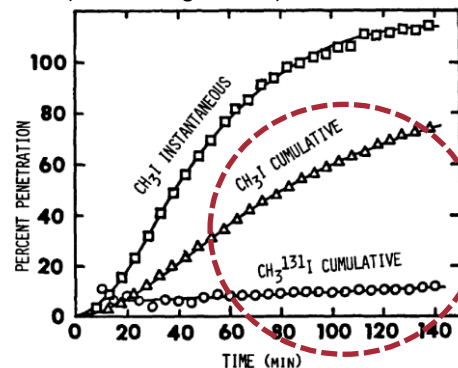
- Significant decrease of available microporosity due to competitive adsorption between H_2O and CH_3I (micropore filling due to H_2O : about 90%) → decreasing physisorption → significant decrease of DF
- Effect of isotopic exchange → determining mechanism for $\text{CH}_3^{131}\text{I}$ trapping → increasing DF with KI impregnation

Effect of isotopic exchange

- cannot be observed at low R.H.
- observable at high R.H. but with poor AC performance



Effect of isotopic exchange in the literature * (breakthrough curves)



Significant effect of isotopic exchange in the breakthrough phase

Previous DF measurements:

- Low R.H. (40%) → retention phase → not observable
- High R.H. (90%) → breakthrough phase → observable

Necessity to determine the breakthrough curves of $\text{CH}_3^{131}\text{I}$ for KI impregnated ACs in order to better explicit the effect of isotopic exchange

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Breakthrough curves measurements

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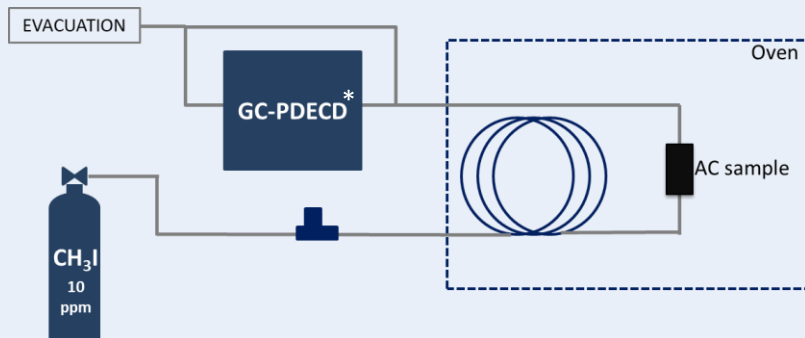
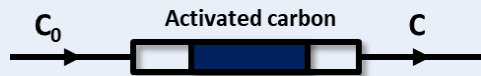
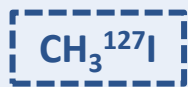
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Breakthrough curves measurements of $\text{CH}_3^{131}\text{I}$ and $\text{CH}_3^{127}\text{I}$

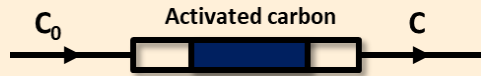
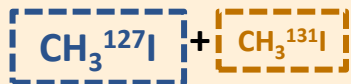
Configuration 1



Two configurations with the same operating conditions

- $T = 20 - 30 \text{ }^\circ\text{C}$
- R.H. = 0%
- Grain size: 1 – 1.4 mm
- $C_0 = 10 \text{ ppm}$
- ^{131}I activity = 10 MBq
- Residence time = 0.2s
- Linear velocity = 25 cm/s

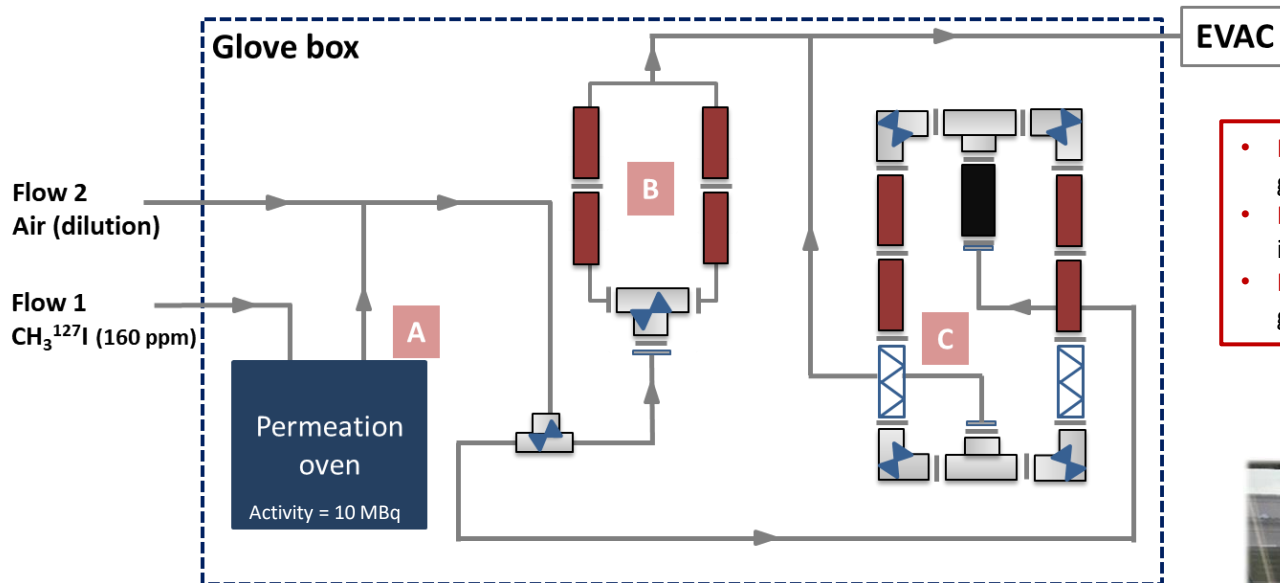
Configuration 2



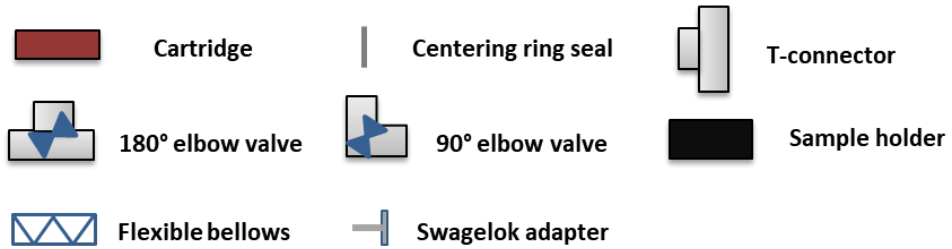
Development of a new experimental set-up

- Continuous generation of stable γ -labelled methyl iodide
- Characterization of obtained γ -labelled methyl iodide flux
- Easy manipulation for data acquisitions
- adapted measuring technique
- Compact size for the glove box

Breakthrough curves measurements of $\text{CH}_3^{131}\text{I}$ and $\text{CH}_3^{127}\text{I}$

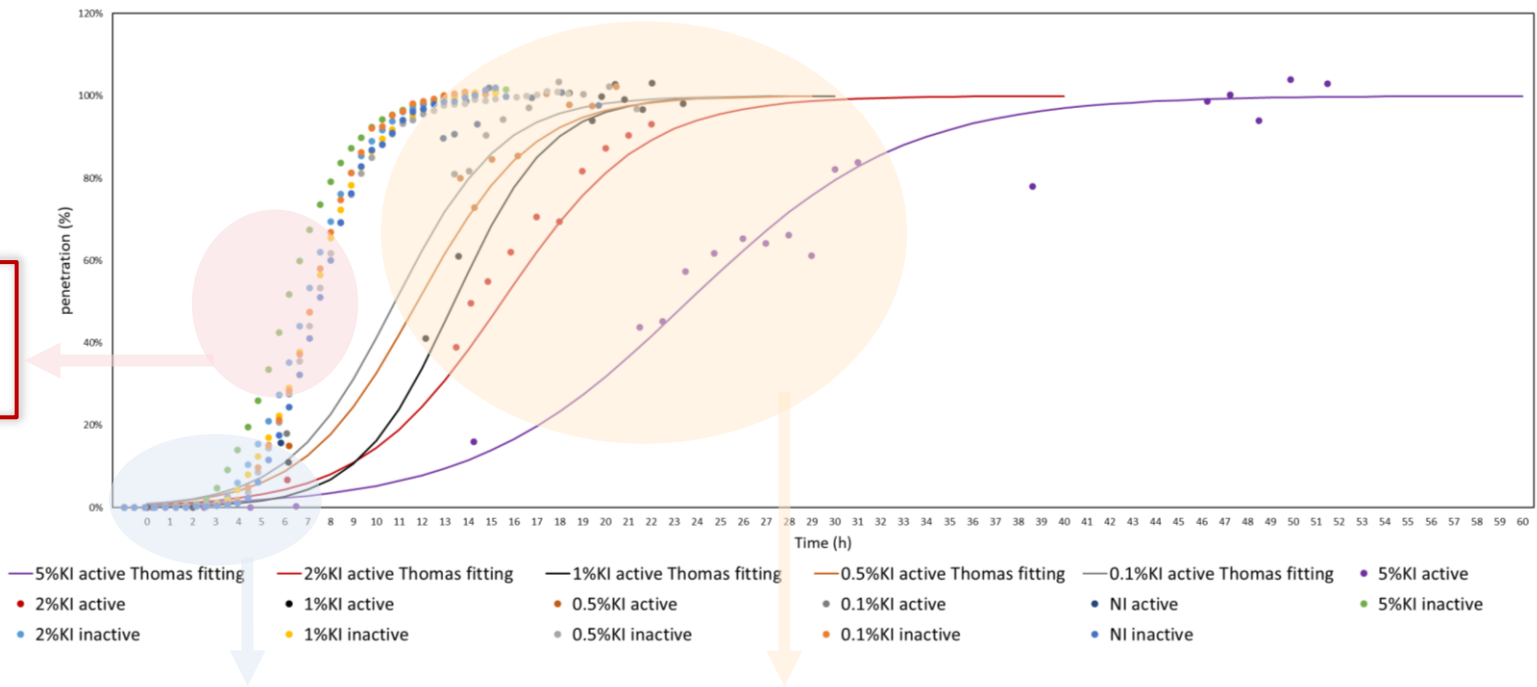


- **Part A:** generation of γ -labelled methyl iodide in gaseous form *via* permeation oven
- **Part B:** characterization of the γ -labelled methyl iodide flux (*ex-situ*, gamma spectroscopy)
- **Part C:** Breakthrough curve measurement (*ex-situ*, gamma spectroscopy)



Breakthrough curves measurements of $\text{CH}_3^{131}\text{I}$ and $\text{CH}_3^{127}\text{I}$

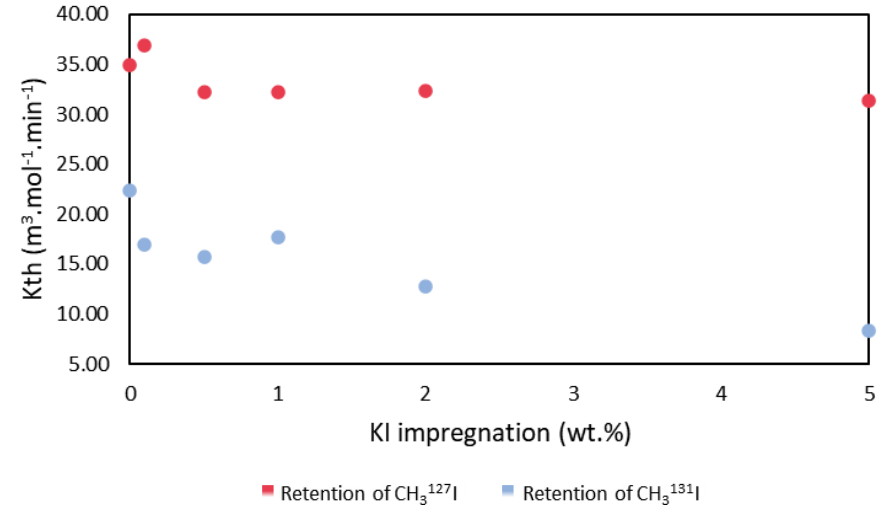
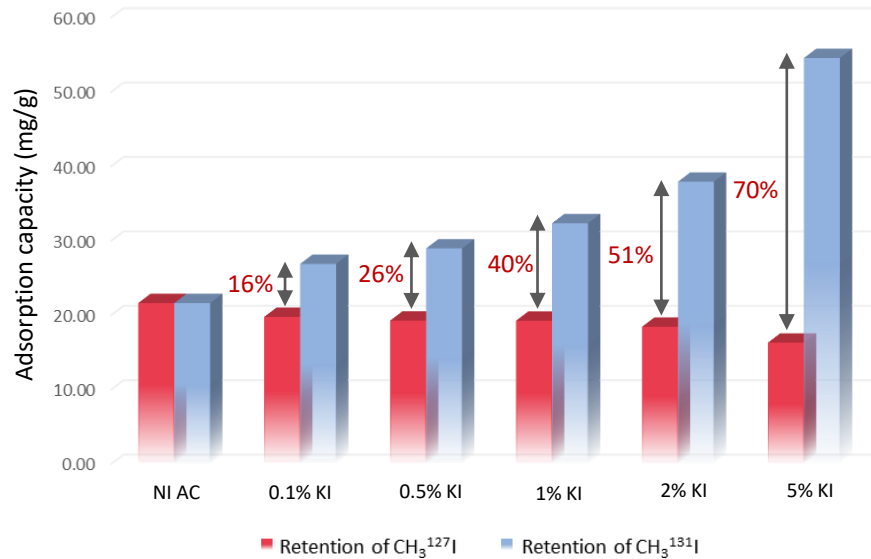
• Quasi – similar breakthrough curves for the trapping of $\text{CH}_3^{127}\text{I}$



• No significant change in the retention phase (in agreement with literature and previous DF measurement tests)

• Significant increase of the performance regarding the trapping of $\text{CH}_3^{131}\text{I}$ due to isotopic exchange during breakthrough phase

Breakthrough curves measurements of $\text{CH}_3^{131}\text{I}$ and $\text{CH}_3^{127}\text{I}$



- Slight decrease in adsorption capacity of $\text{CH}_3^{127}\text{I}$ due to the slight increase of the mass of the AC sample
- Progressive increase in adsorption capacity $\text{CH}_3^{131}\text{I}$ due to the effect of isotope exchange (gain of about 70% for 5wt.% in KI)
- Significant decrease of the rate constant K_{th} due to the isotopic exchange (increase of adsorption sites participating in the isotopic exchange)

Conclusions and perspectives

AC performance towards the trapping of $\text{CH}_3^{131}\text{I}$ (T = 20 -30 °C)

DF measurements

- Good performance of all the tested ACs at R.H. = 40% (DF: 10^4 - 10^5)
- Drastic decrease of DF at R.H. = 90% (maximum DF around 100)
- The best performances are observed for impregnation compositions similar to nuclear grade AC
- DF decreases with KI impregnation at R.H. = 40% → absence of the isotopic exchange for the KI impregnated AC at R.H. = 40%, with the AC performance dominated by physisorption and pre-adsorbed H_2O
- DF increases with KI impregnation at R.H. = 90% → Observation of the isotopic exchange

Breakthrough curve measurements (effect of KI)

- Quasi – similar breakthrough curves for the trapping of $\text{CH}_3^{127}\text{I}$
- No significant change in the retention phase (in agreement with literature and previous DF measurement tests)
- Significant increase of the AC performance for the trapping of $\text{CH}_3^{131}\text{I}$ due to the isotopic exchange (during the breakthrough phase)

Perspectives

- Continue the breakthrough curve measurements with different ACs (TEDA and TEDA/KI co-impregnated ACs)
- Improvements of the newly designed experimental set-up (temperature, R.H., etc)
- Investigations of the effect due to isotopic exchange phenomena for other synthesized materials

THANK YOU FOR YOUR ATTENTION.