

# **Gas Purification: Goal & Status**



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# Goals in terms of background

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- Crucial to be able to identify the contaminants
  - ▶ Traces Uranium, Thorium, Oxygen, H<sub>2</sub>O, Radon, Krypton etc.
- Oxygen, H<sub>2</sub>O, electronegative impurities
  - ▶ Well removed by hot getter included in the loop system
  - ▶ But radon rate increased: emanates from the getter!
- Radon ...
  - ▶ ~ 50mBq in the sphere at Queen's S30
  - ▶ Level required for Ne mixture from simulations < 48 μBq
- Krypton ...
  - ▶ For R2D2: removing the Kr85 from the Xenon
  - ▶ Level required from simulations < ?

# Acceptable level of radon

- Estimated by simulations: (credits Alexis)
  - ▶  $10^4$  decays of  $^{222}\text{Rn}$  homogeneously distributed in volume
  - ▶  $10^4$  decay of  $^{218}\text{Po}$  /  $^{214}\text{Pb}$  on the inner surface
- Results in dru/Bq...

	He mixture	Ne mixture
$^{218}\text{Po}$	2411	612
$^{214}\text{Pb}$	663	227
$^{214}\text{Bi} + ^{214}\text{Po}$	987	210
Total	4061	1050
To obtain 0.05 dru < 1keV	< $12\mu\text{Bq}$	< $48\mu\text{Bq}$



# Radon mitigation: Strategies

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- Radon trap
  - ▶ Feasible but CH<sub>4</sub> is also absorbed
  - ▶ Tests need to be done to find the optimal conditions of the column (Temperature, Flow) to remove the radon only.  
--> In progress led by José in Marseille.
- Queen's tests and plan with S30
  - ▶ Run plan already in place in order to control the CH<sub>4</sub> amount.
  - ▶ Procedure in place for running with the trap.
- Material for trapping
  - ▶ Carboxen 564: Material also with the lowest radioactivity
  - ▶ Alternative Carboxen 1000



# Radon adsorption measurements at CPPM

Radon : noble gas => physisorption on microporous materials

❖ **Optimum capture** => porous radius  $\approx 2 \times$  Rn atomic diameter

Experimentally : *Porous radius  $\in [0.5 \text{ nm} - 0.7 \text{ nm}]$*

(Capture ability is also enhanced by chemical composition, porous shape, ... )

Many microporous adsorbents : Active charcoals, Carbon Molecular Sieves, Zeolite, Metal Organic Framework, Organic Aerogels, Cryptophanes, etc, ...

❖ **Adsorption competition**

*(Atomic diameter)*

He -> 0.218 nm	Ne -> 0.275 nm
Ar -> 0.340	Kr -> 0.369
Xe -> 0.410	<b>Rn -&gt; 0.417</b>
CH <sub>4</sub> -> 0.380	N <sub>2</sub> -> 0.364

High difficulty to remove Rn from Xe

*Swing adsorption (P,T,V) in optimized adsorbent could be a solution ?*

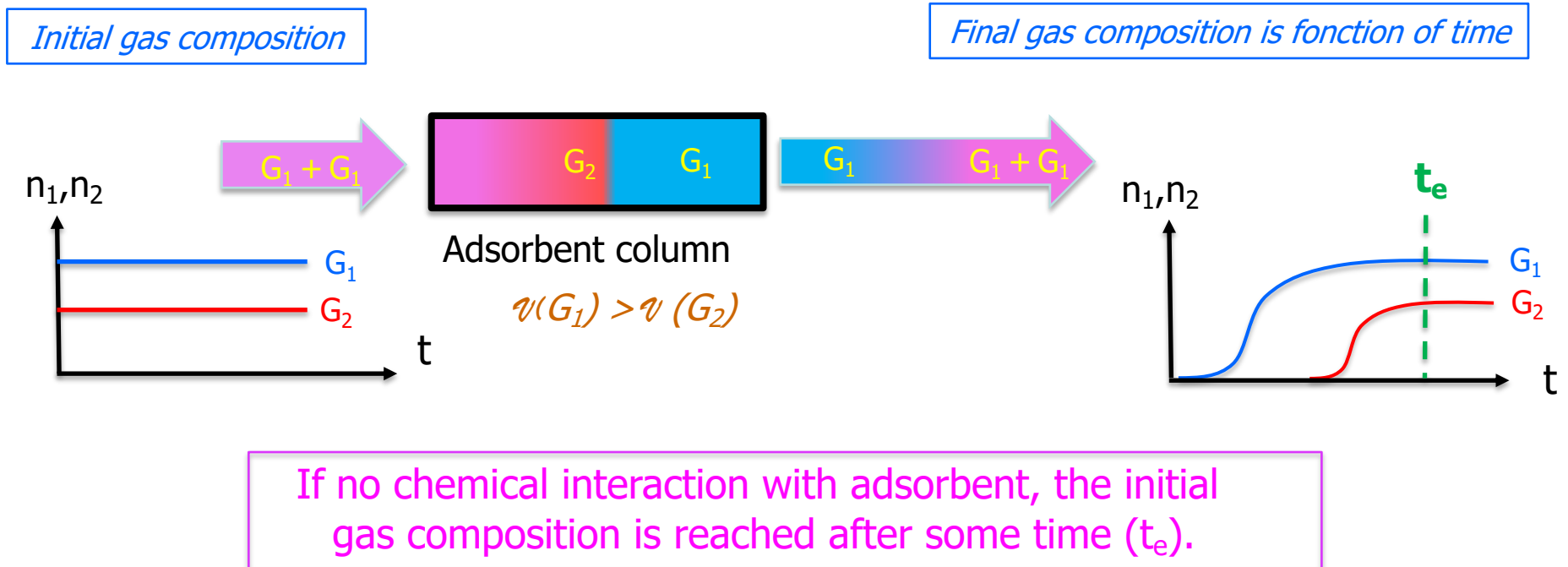
## ❖ Radon capture in mixed gases with continuous circulation

In a chromatographic column each gas component has different velocities



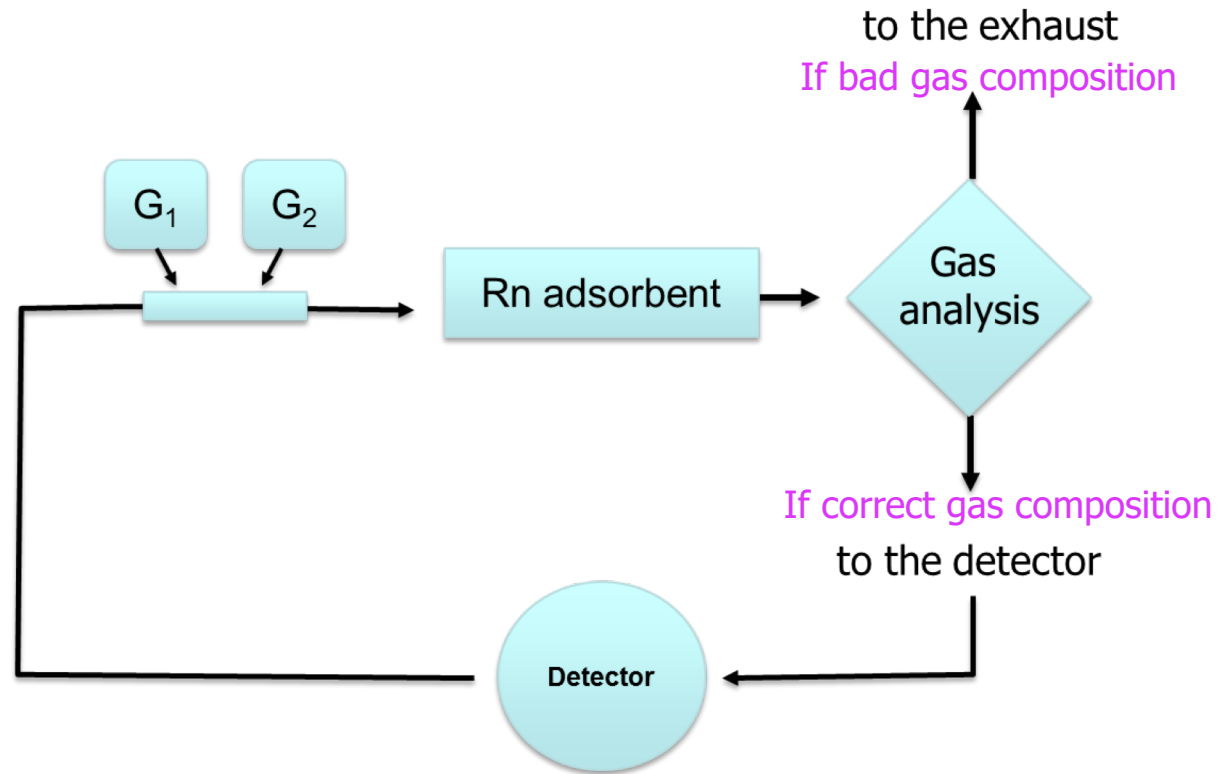
The initial gas composition is destroyed (for some time)

(He + CH<sub>4</sub>, He + Ar, Ar + CH<sub>4</sub>, Ne + CH<sub>4</sub>, CF<sub>4</sub> + CHF<sub>3</sub> + C<sub>4</sub>H<sub>10</sub>)



We need optimization of adsorbent to enhance Rn capture, and reduce the delay time

## Basic setup

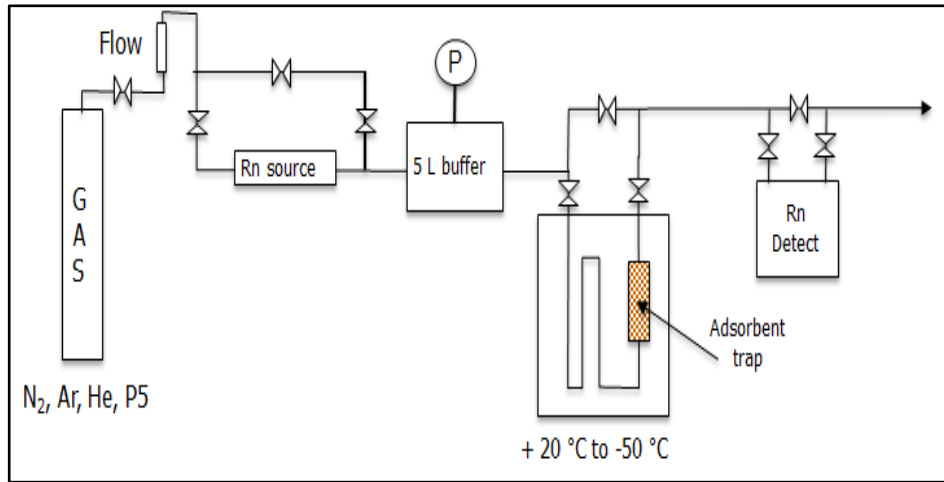


**Gas analysis** : many technologies (IR, UV, catality, semi-conductor, ...)

Ex : OLDHAM OLCT 100 – XP- IR  
Range 0 – 5 %  $CH_4$

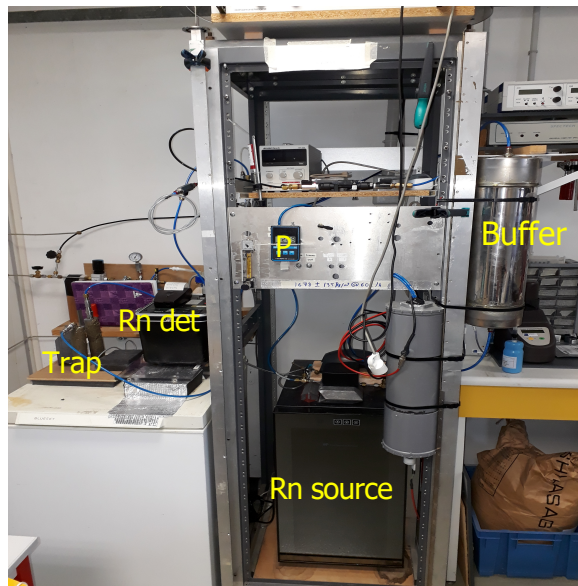


## ❖ Radon adsorption test bench of CPPM



$$\text{Rn adsorption} \left[ \frac{\text{m}^3}{\text{kg}} \right] = \frac{\text{Rn activity in adsorbent}}{\text{Rn activity in gas}}$$

Activity in adsorbent => Ge spectrometry  
Activity in gas => Lucas Cell or RAD7



### Performed measurements

- 60 adsorbents samples measured @ 20, 0, -30, -50, -80 °C
- Carrier gases used N<sub>2</sub>, He, Ar,
- Collaboration with physical-chemist  
=> *microscopic adsorbent properties*

### Current tests and projets

- Rn adsorption in Ar + CH<sub>4</sub>, Ne + CH<sub>4</sub> -> Marie Cécile
- Rn adsorption in CF<sub>4</sub>+CFH<sub>3</sub>+Isobut -> MIMAC (Daniel)
- Rn adsorption in Xe -> R2D2 (big challenge)

For special gases (Ne, Xe, CF<sub>4</sub>,...), a closed circuit is required

# Summary

- Radon can be capture in microporous adsorbents with high efficiency
- Competition between Rn and carrier gas need new optimized materials
- Cleaning of gas mixture is possible but we need a more in-depth study.
- A radon adsorption facility exist at CPPM (Marseille) for optimization of radon capture in different gases and materials.
  - Several studies in progress or in project

# Radon mitigation: Strategies

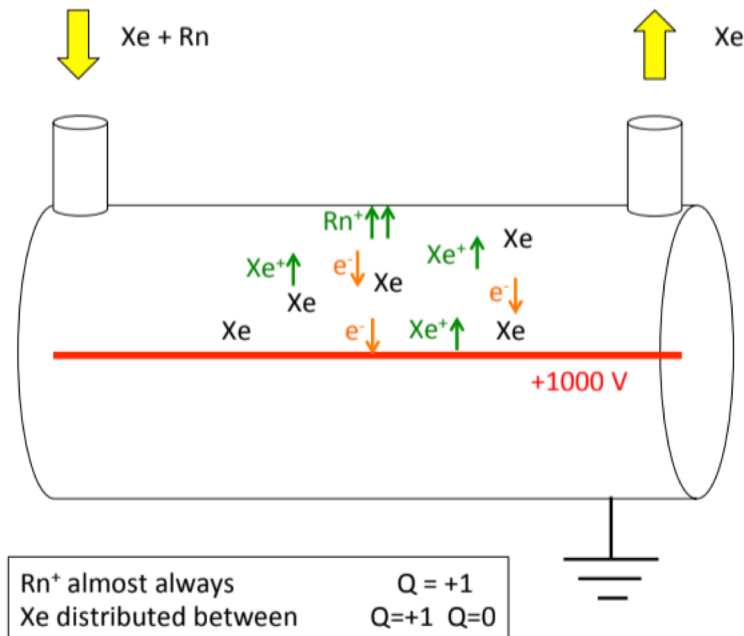
- Electrophoretic radon removal
  - ▶ Based on the first ionisation energy, exploiting favorable ion charge-exchange dynamics.

	First ionization Energy (eV)
Rn	10.4875
Xe	12.14
Ne	21.56
CH <sub>4</sub>	12.61
F	17.42
Ar	15.75
C <sub>3</sub> F <sub>8</sub>	13.38

- ▶ By comparing the energy, in collision with xenon ions, radon will be efficiently ionized via charge transfer:  $\text{Xe}^+ + \text{Rn} \rightarrow \text{Xe} + \text{Rn}^+$

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# Distillation

## Thermodynamic properties:

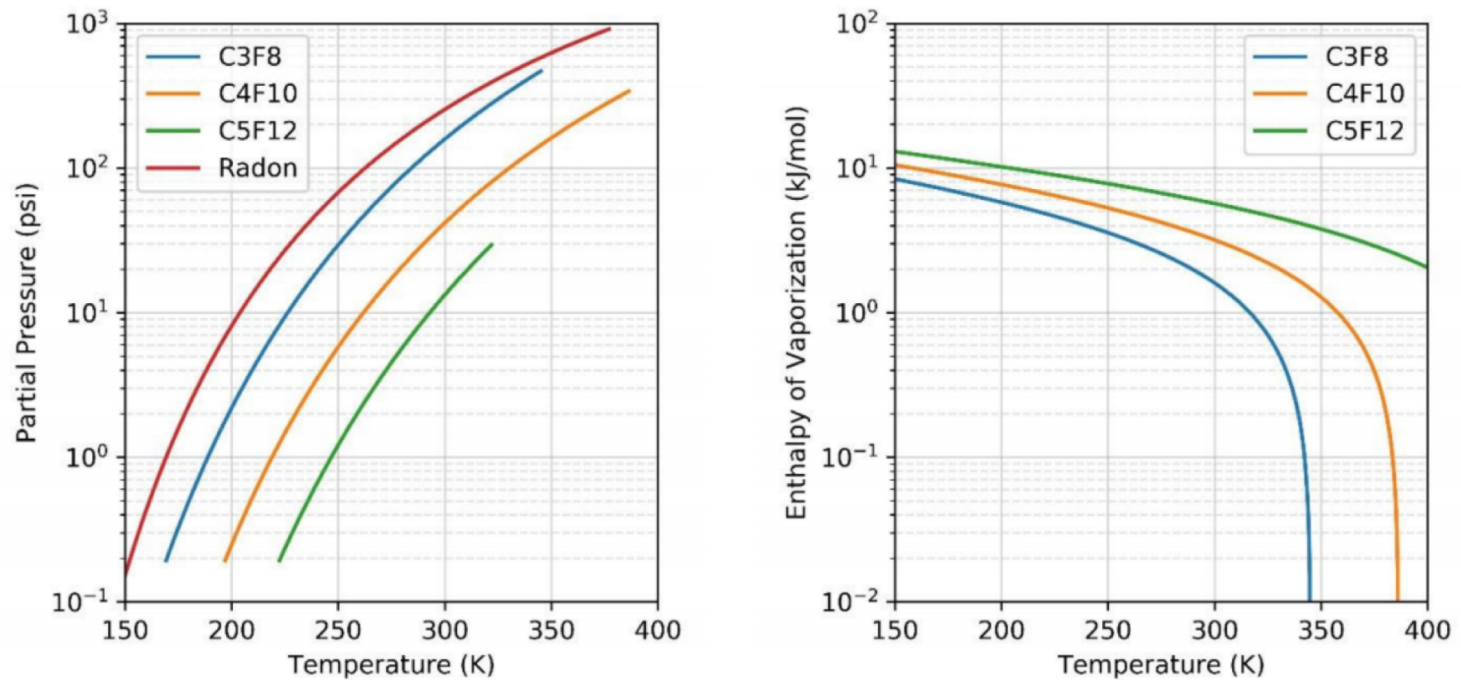
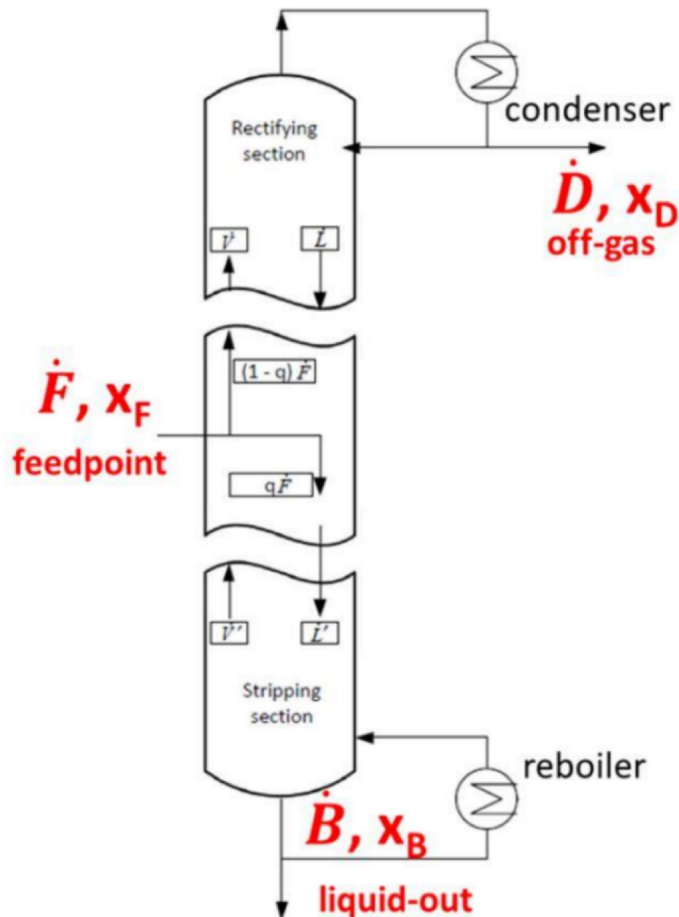


Figure 1: (left) Vapour pressures of Radon and fluorocarbons against temperature and (right) enthalpy of vaporization at saturation of fluorocarbons against temperature.



# Distillation



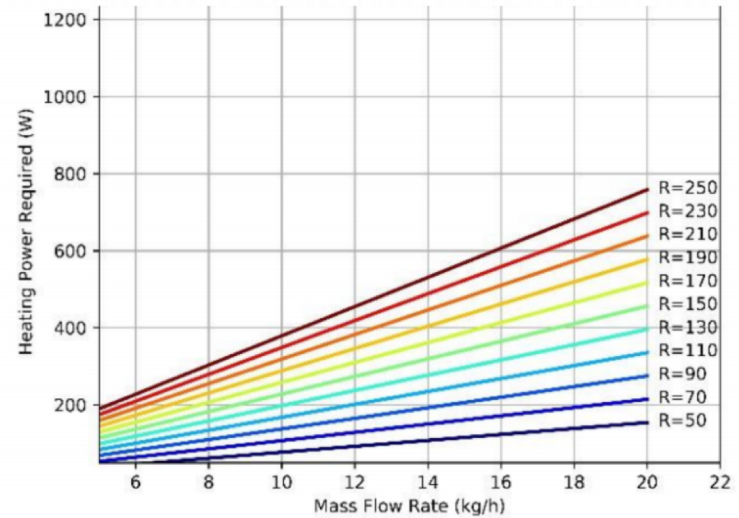
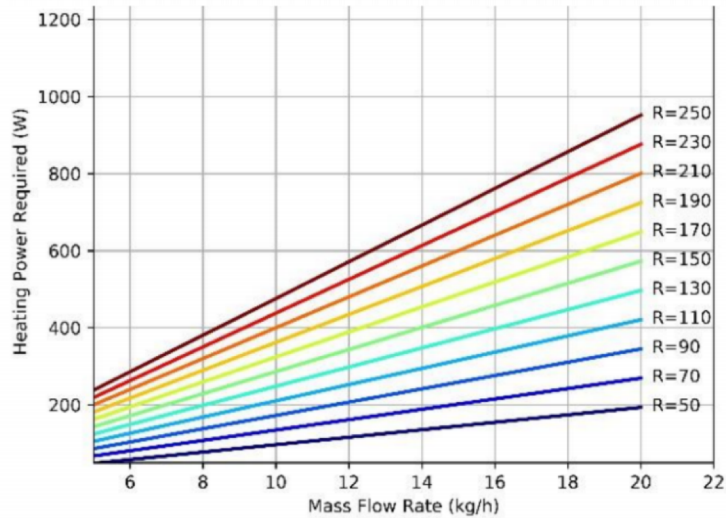
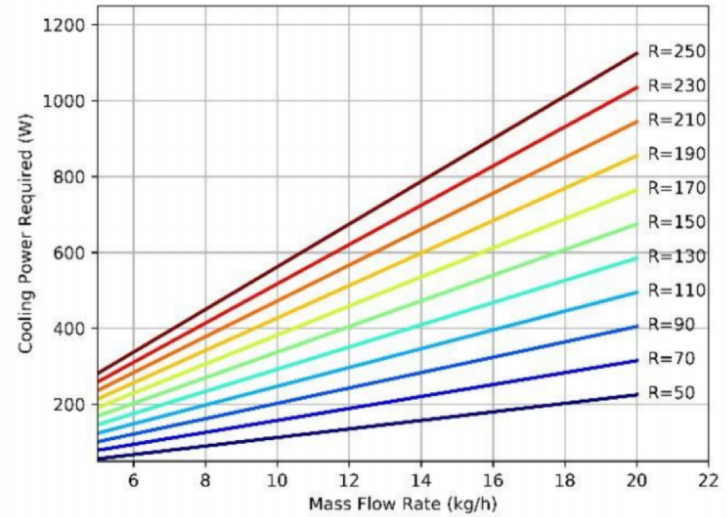
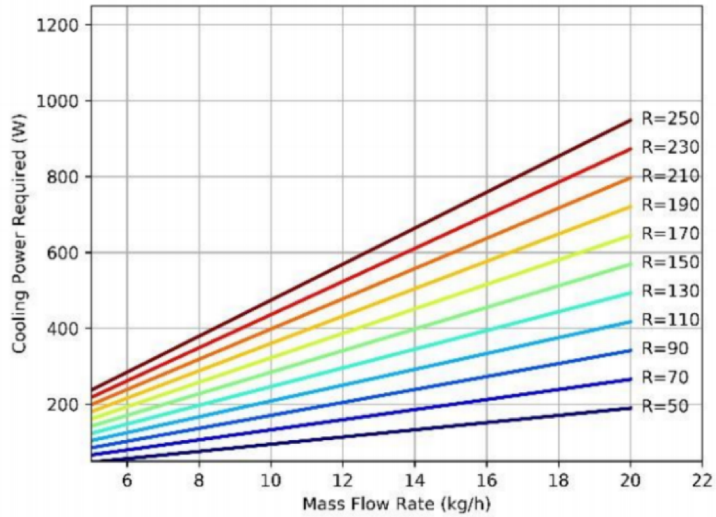
The required heating power and the required cooling power depend on:

- the vapor flow from the stripping (bottom) section
- the liquid flow from the rectifying (top) section
- the enthalpy of vaporization, plotted for different temperatures

These calculations assume a gas feed into the column at:

- saturation temperature and pressure
- no heat loss to the surroundings
- flow rate into the column
- the reflux ratio

# Distillation



# Distillation

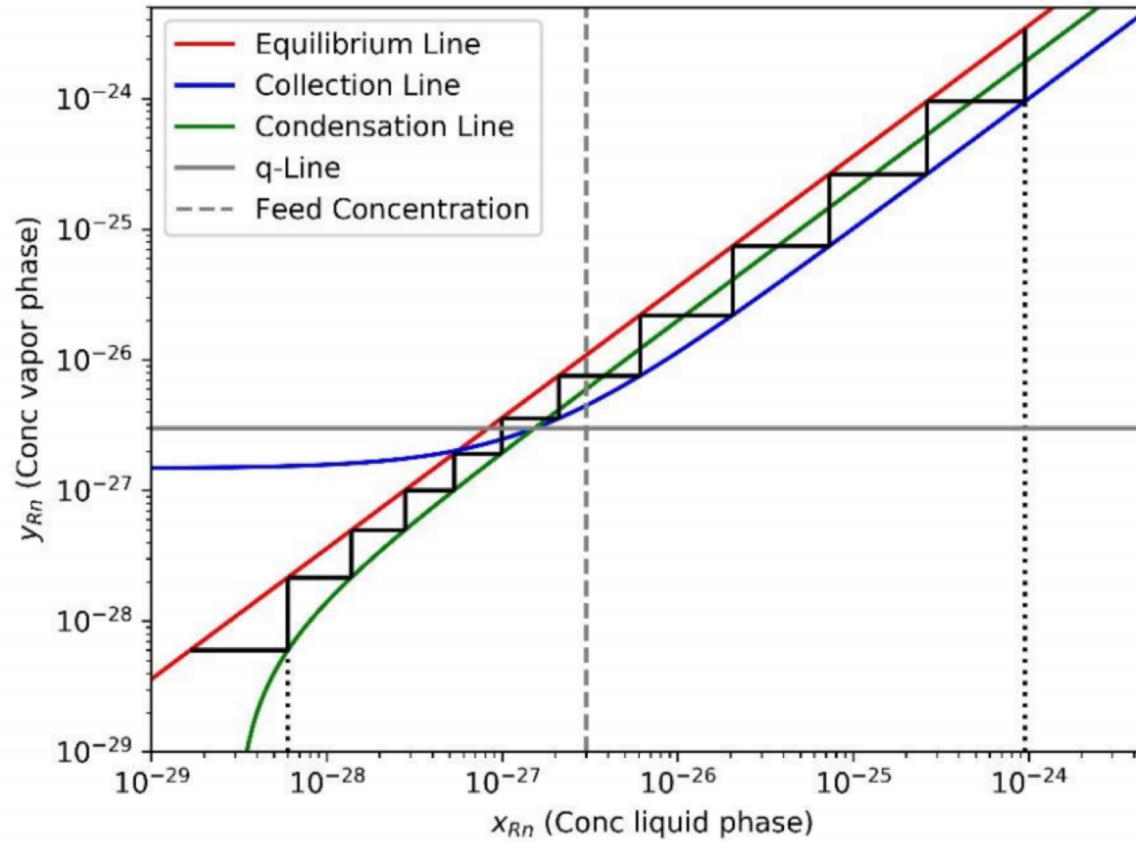


Figure 5: McCabe Thiele diagram for distillation of Radon from  $C_4F_{10}$ , with a gaseous feed, and a reflux ratio of 200. The number of theoretical stages is 11.



# Distillation

Table 3: Possible distillation column parameters for gaseous feeds of  $C_3F_8$  and for  $C_4F_{10}$  at 200 psi.

<b>Parameter</b>	<b><math>C_3F_8</math></b>	<b><math>C_4F_{10}</math></b>
Reflux Ratio	350	150
Reduction Factor	50	50
Mass Flow Rate	6 kg/h	12 kg/h
Minimum Column Height	1.29 m	0.35 m
Heating Power Required	288.4 W	93.4 W
Cooling Power Required	402.2 W	274.6 W

Table 4: Possible distillation column parameters for liquid feeds of  $C_3F_8$  and for  $C_4F_{10}$  at 200 psi.

<b>Parameter</b>	<b><math>C_3F_8</math></b>	<b><math>C_4F_{10}</math></b>
Reflux Ratio	300	75
Reduction Factor	50	50
Mass Flow Rate	6 kg/h	12 kg/h
Minimum Column Height	1.11 m	0.39 m
Heating Power Required	345.9 W	139.1 W
Cooling Power Required	344.7 W	137.3 W



# Distillation

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- **Summary:**

- Simulations with McCabe method done
- Exploring stripping column to improve factor reduction
- Compare the 2 models
- CFI funded to build the column
- Start this summer

- **Goal and time lines:**

- Distillation ready by December 2019/ beginning of next year
- Paper to be published with the code available