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Belgian Nuclear Research Centre

Measuring diffusion of gas in partly saturated clay

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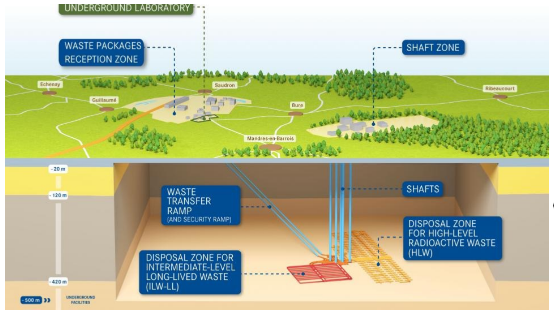
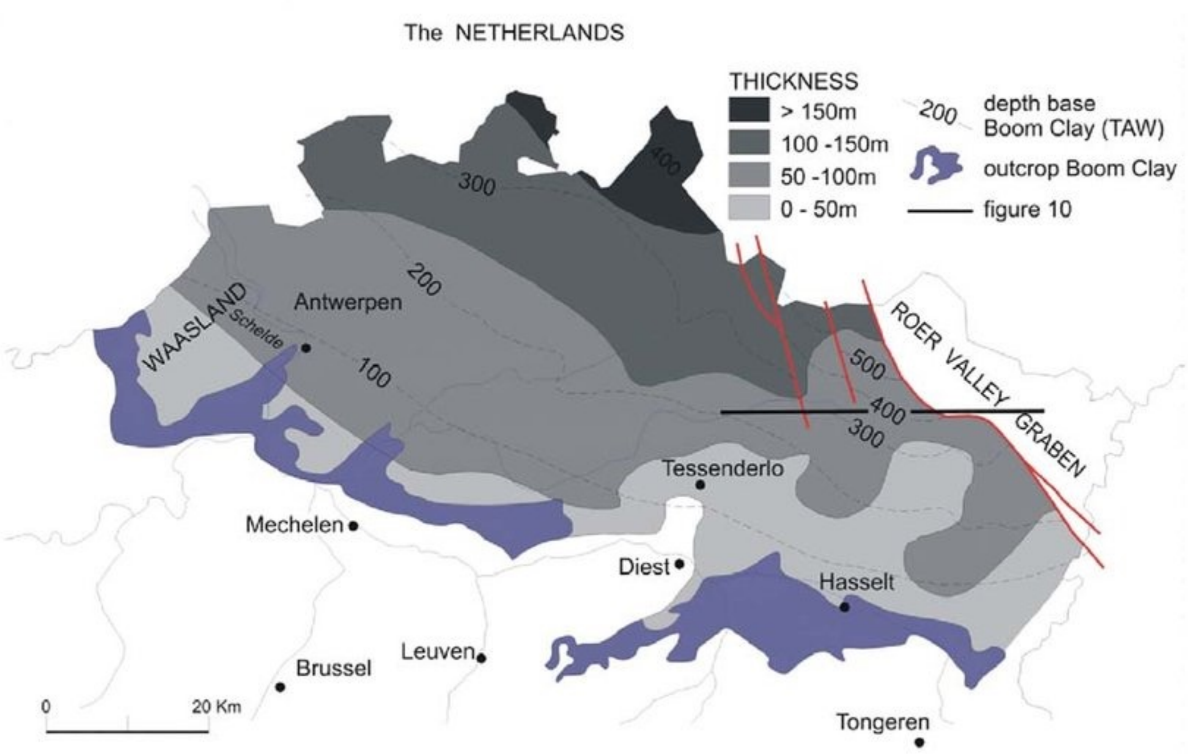
A bit of background



A bit of background

- Disposal in clay
- Why clay?
- Low hydraulic conductivity
- High sorption capacity
- Self sealing

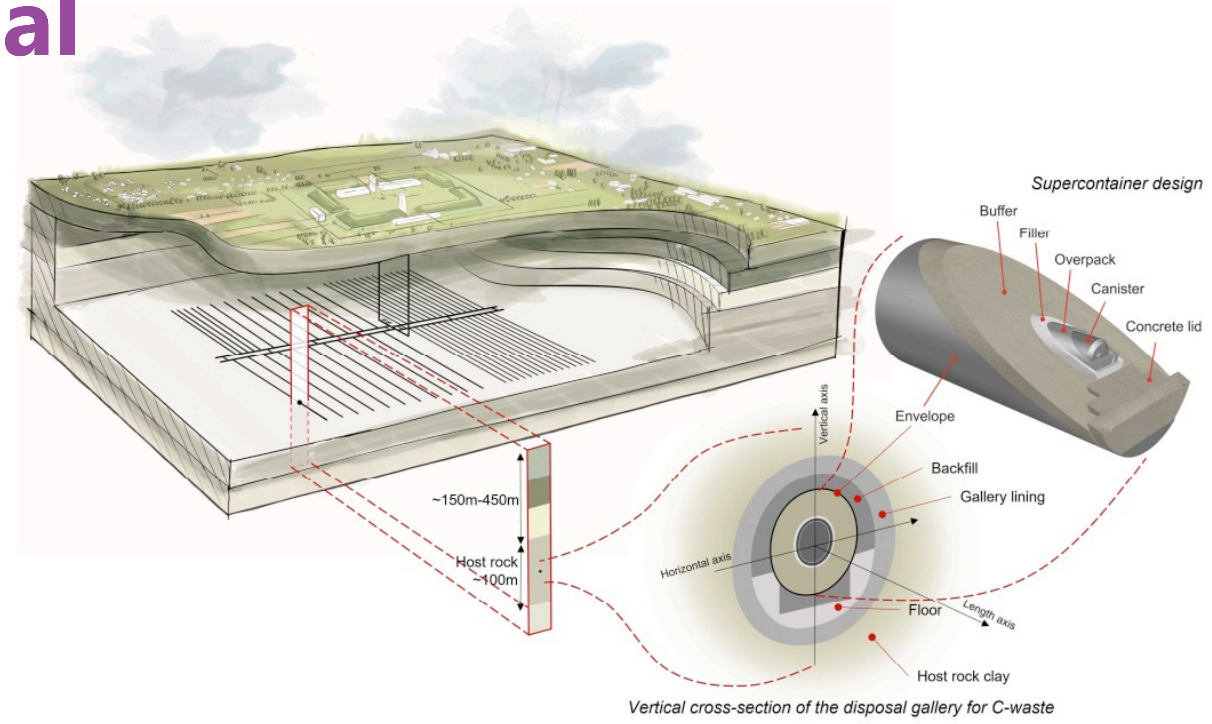
- Reduced mobility of radionuclides



Potential host rock in Belgium: Boom Clay

Schematic view of the current design of geological disposal facility in Belgium, (ONDRAF/NIRAS)

Gas in geological disposal



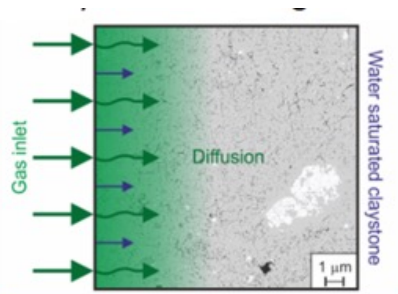
- Considerable amounts of gas can be produced in a repository
  - Mostly hydrogen (anaerobic corrosion of steel and reactive metals)
  - Degradation of organics wastes & radiolysis also produce gas

→ Significant volumes: several times the excavated volume but generated over a long time period

→ Different evacuation pathways

Gas transport modes in clays

- Transport of solutes → mainly diffusion
  - Characterized by diffusion coefficients
- Need for diffusion coefficients:
  - Radionuclides
    - Many data available
  - Dissolved gases (gas generation in repository)
    - Noble gases, light hydrocarbons, hydrogen

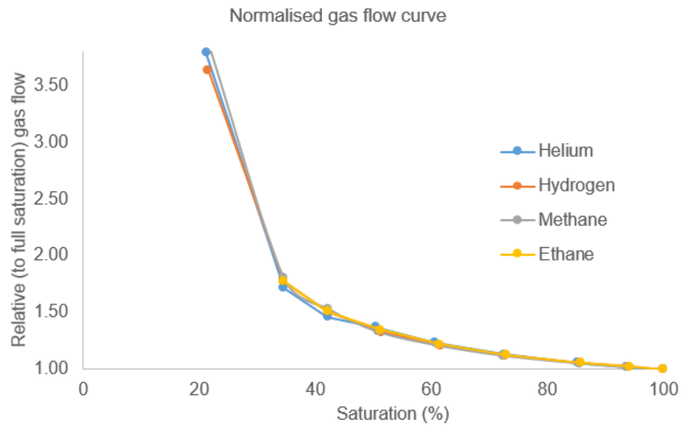


diffusive transport of dissolved gas

$$J = -D\frac{dC}{dx}$$

D is diffusion coefficient
C is concentration
x is distance
J is flux

Open questions

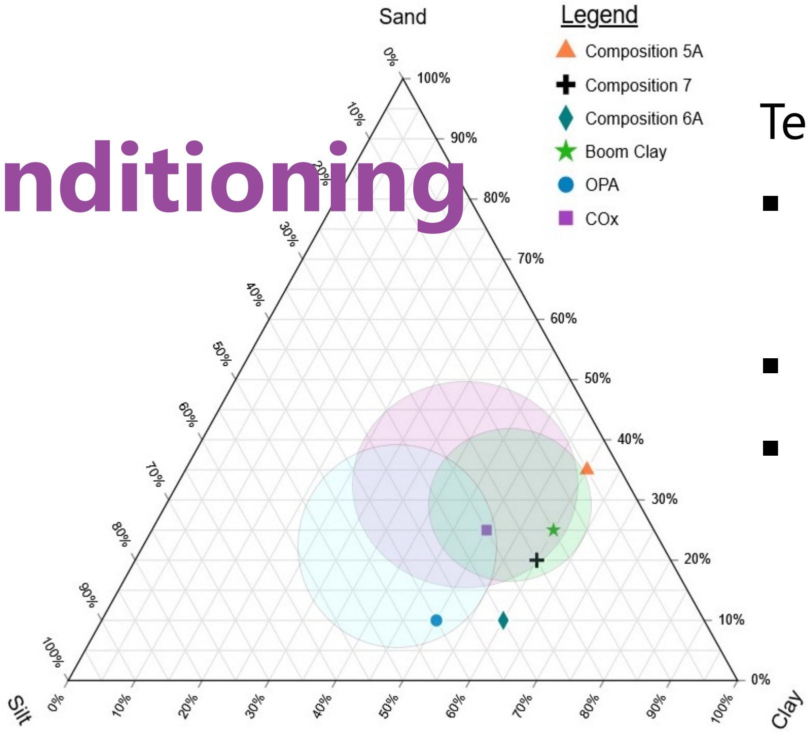


- What we have?
  - Large data set of diffusion coefficients
    - Different gases (noble gases, light hydrocarbons, H_2)
    - Different materials (Boom Clay, Sand of Eigenbilzen, Callovo-Oxfordian clay, Opalinus Clay, bentonite, shale, cement-based materials, geopolymers, …)
    - But: all on saturated samples

- What is missing?
  - Unsaturated conditions might exist in host rock and engineered barriers
    - After repository closure or due to gas generation
    - What is impact of desaturation on gas diffusion coefficients?
    - Also need to understand process
      - Use “simple” systems with known mineral phases → easier to understand + transferability of results

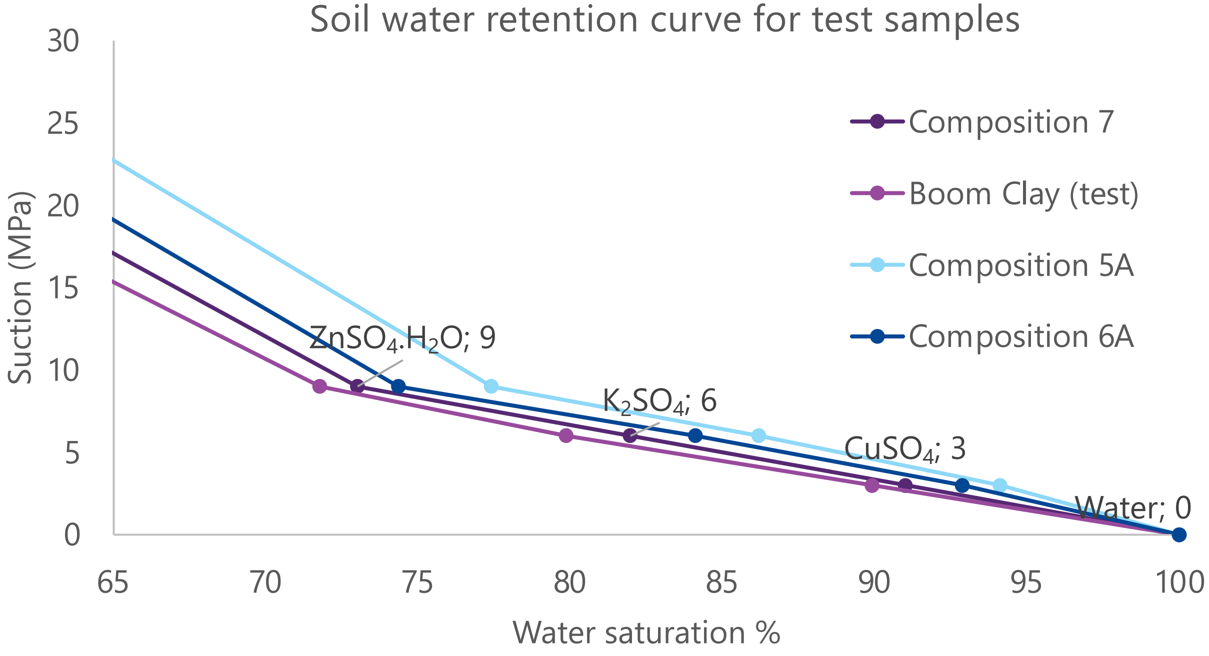
Samples & conditioning

Synthetic clay sample conditioning – vapour equilibrium.

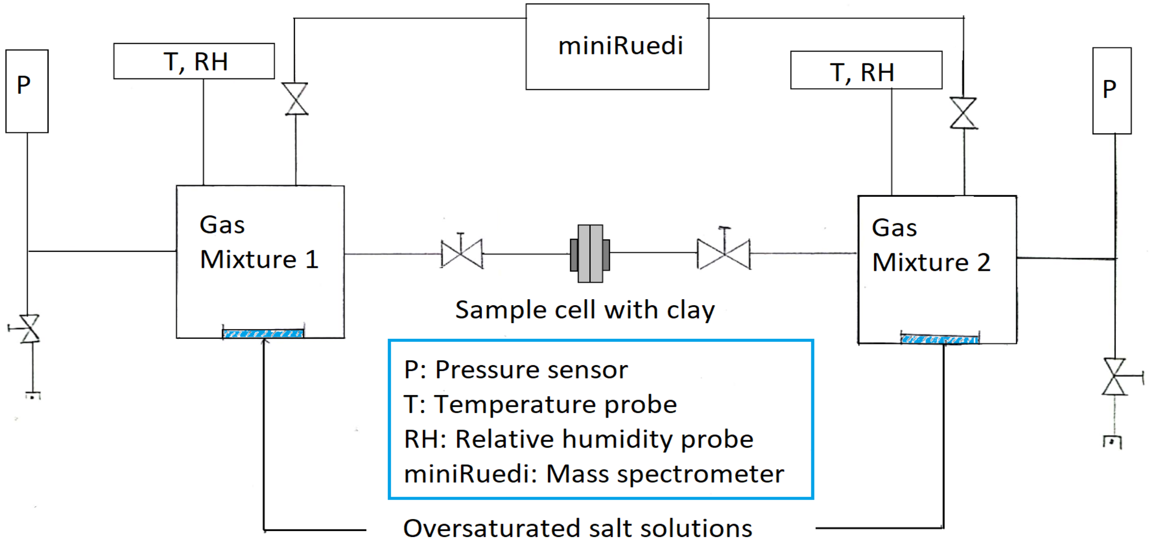


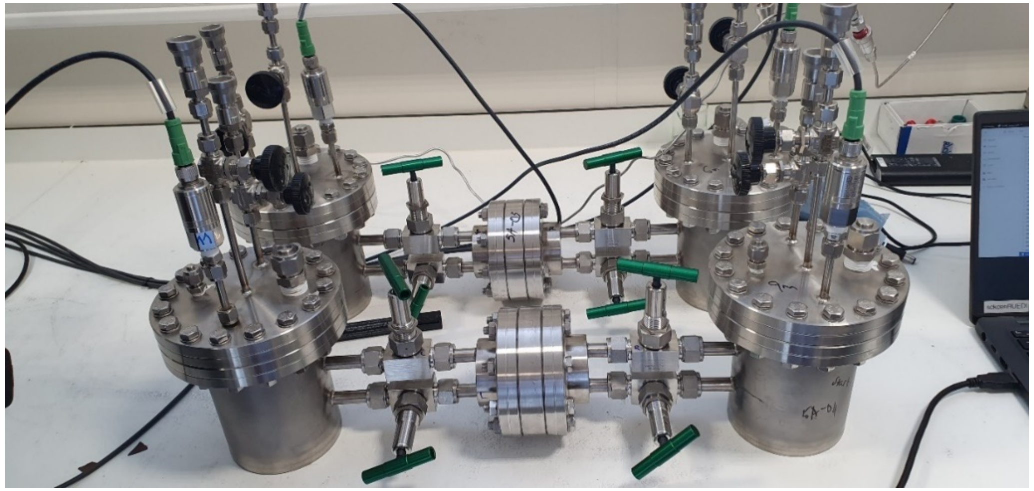
Ternary mixtures of sand, silt and clay.

- Clay : 80% MX80 and 20% kaolinite (63-125 µm)
- Sand: Pure quartz sand (63-125 µm)
- Silt: Pure muscovite mica (<63µm).



Overview- Diffusion experiments





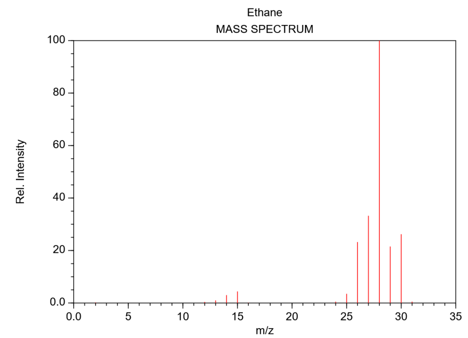
- Equal pressure in both vessels (around ambient) → diffusion only
- Room temperature
- Constant RH by using oversaturated salt solutions
- Measure gas concentration increase as function of time

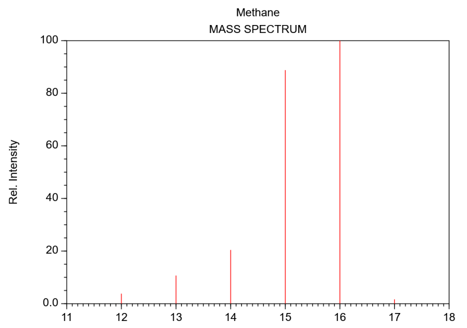
Gas analysis

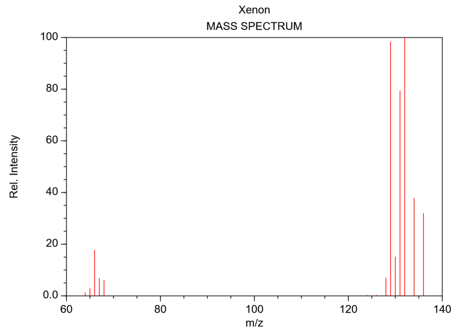
- First idea: cross diffusion of 2x 2 gases
  - He and CH_4
  - Xe and C_2H_6
  - → too much peak overlap

- Simplified idea:
  - He
  - Ar
  - → no more overlap (but less data per test)

	m/z	Detector
He	4	F
CH_4	15	F
Xe	129	F
C_2H_6	30	F
Ar	40	F



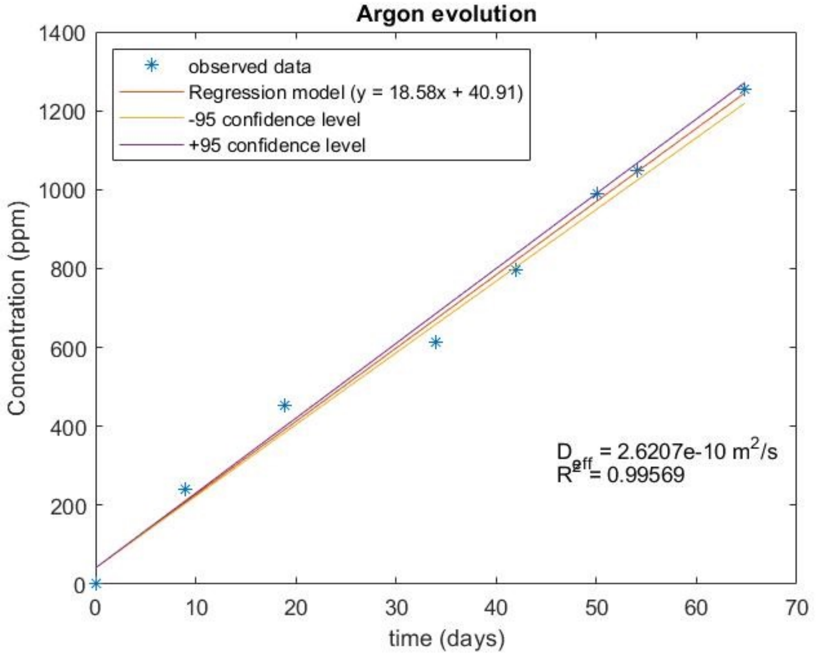




Gas analysis

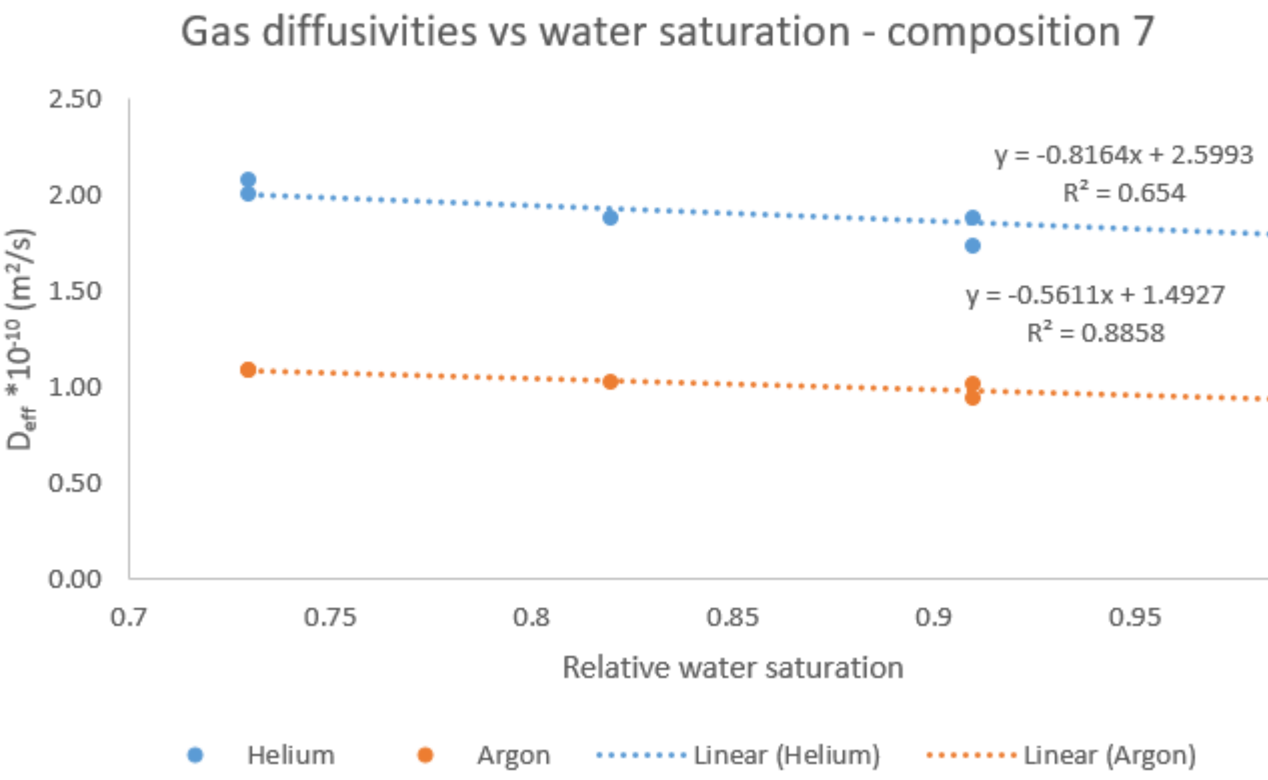
- Effective diffusivity- analytical solution of Fick's first law

$$N_A = \left[\frac{D_{AB} P_t}{RTZ}\right] \ln\left[\frac{1 - y_{A2}}{1 - y_{A1}}\right]$$



Results

- Series of diffusion coefficients for synthetic samples



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