KaMu

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The influence of environmental conditions on gas generation in the disposal of LLW KaMu (2021-2022)

VTT

Aim: To create disturbances in the gas generation experiment and study the influences on the gas generation, water chemistry and microbial activity.

- The addition of sulphate (simulating flow of sulphate containing groundwater to the repository)
- The influence of high(er) pH (simulating the influence of concreate environment)
- The utilization of the results on modelling – verification of GRM (The generalized Repository Model)

Gas Generation Experiment (GGE)

- *In situ* large-scale experiment in Olkiluoto repository operated by TVO (1997→)
- 16 waste drums (200 L) were filled with LLW maintenance waste, placed in a concrete box and closed in the gas tight tank of acid proof steel (20 m^3)
- The tank was filled with river water
- Temperature is maintained in the level of +8°C -+11°C
- No mechanical stirring of the water in the tank.
- Analyses : Gas generation, composition of gas, water chemistry, key microbial groups

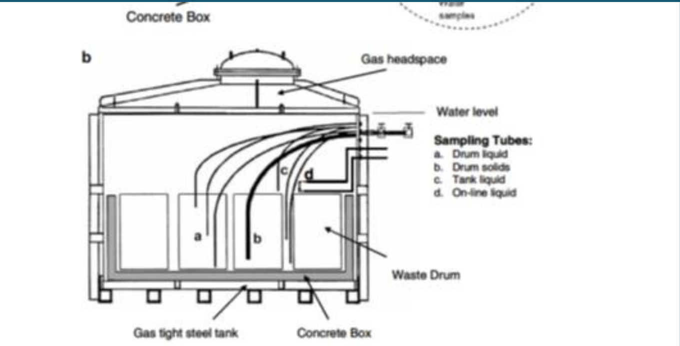

Fig. 1. Plan (a) and side elevation (b) of the gas generation experiment showing the position of the waste drums and sampling lines.

Small et al. 2008

Gas generation in repository conditions

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- Gaseous radionuclides (e.g. ^{14}C) can be transported to the biosphere in the form of methane gas ($^{14}CH_4$)
- Development of overpressure in the repository
- Disruption of the engineered barrier system (EBS)
- Produced gas in the geosphere can enhance the activity of microbial communities
- Enhanced migration of radionuclides in groundwater to the biosphere

1. Biodegradation of organic materials:

$C \dashrightarrow CO_2, CH_4, (H_2)$

2. Corrosion of metals in the waste and packaging (drums)

$Fe + 2\,H_2O \rightarrow Fe(OH)_2 + H_2$

3. Radiolysis of water and some organic molecules in the waste packages, generating mainly hydrogen

Cellulose and hemicellulose are easily biodegradable

Gas Generation

- Gas generation started one year after the initiation of the experiment
- Gas is composed mainly CH_4 (80-90%) and small concentrations of H_2, N_2, O_2 and CO_2
  - H_2 rapidly consumed by microbes
  - CO_2 → tank water (increased DIC)
- Gas generation rate increased ~2005
  - pH decreased close to neutral also in the drum lid level

Gas generation is assumed to continue at constant level

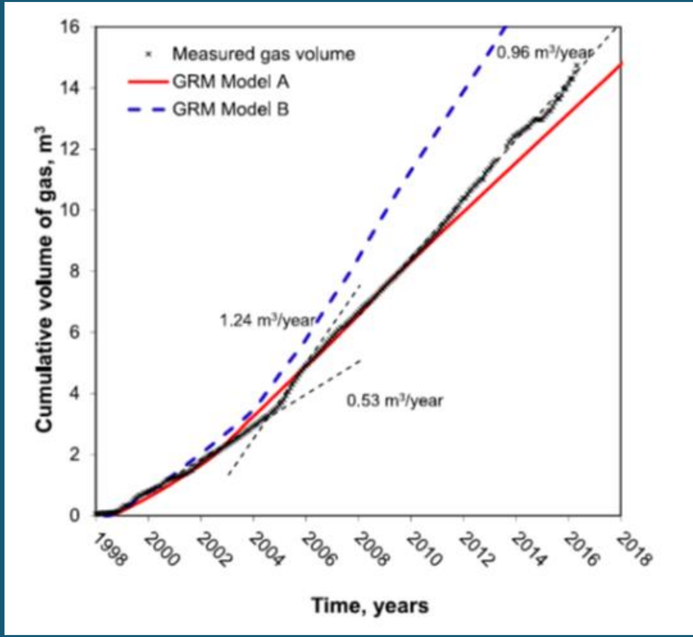
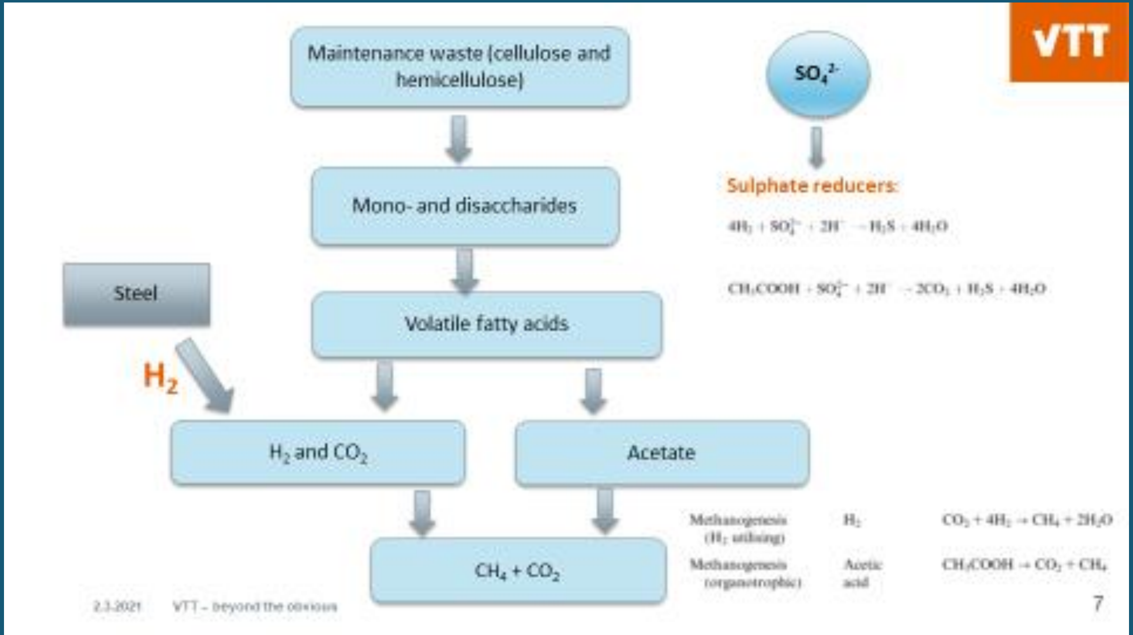


Figure: Small et al. 2017

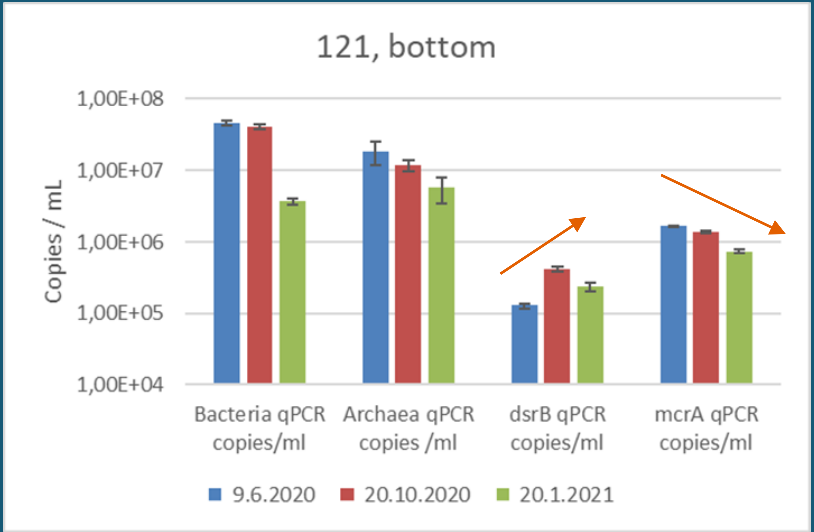
Task 1. Sulphate addition

- Research question:
  - Can sulphate addition enhance activity of sulphate reducers and reduce gas generation?
- Sulphate concentration in the tank water was below detection limit
- Sulphate reducers use sulphate as electron acceptor
- Followed parameters: gas generation, gas composition, key microbes, chemical paramaters.



Task 1. Sulphate addition

- After the addition of K_2SO_4 and mixing of the tank water, sulphate concentration was increased to 100 mg/L
- After SO_4-addition:
  - Small increase in sulphide concentration
  - Increse in the amount of sulphate reducers
  - Decrease in the amount of methanogens
    - Sulphate reduction on-going?
- So far no major influences on gas generation and composition.
- The results will be used to verify GRM modelling.



Sulphate addition ⇩ Mixing ⇩

	9.6.2020	22.6.2020	20.10.2020	18.1.2021
SO_4^{2-}	<0.2	940	102	98
S_2^-	<0.03	<0.03	0.09	0.11

Next steps (Task 2. High pH)

- Concrete has assumed to create alkaline environment in the repository → inhibition of microbial activity
- In the beginning of the experiment chemical conditions were heteregeneous in different compartments of the experiment (pH, DOC)
  - optimal niches for microbial activity
- Microbial activity (CO_2, volatile fatty acids) has neutralised concrete conditioned alkaline water.
- pH will be increased by adding e.g. KOH
  → gas generation, gas compostition, microbiology and chemical paremeters will be monitored

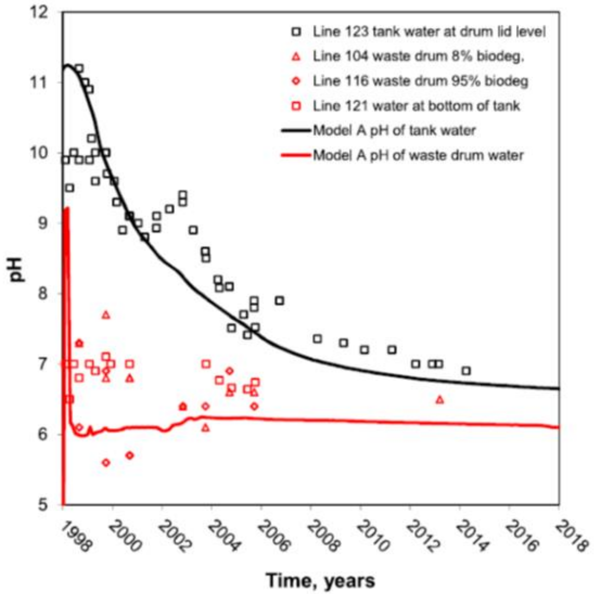


Fig. 7. Measured pH of water samples from sample lines from the GGE and fitted average modelled pH of model cells representing tank water and waste drum regions of the experiment.

Figure: Small et al. 2017

Publications

- Small, J., Nykyri, M., Vikman, M., Itävaara, M., Heikinheimo, L., The biogeochemistry of gas generation from low-level nuclear waste: Modelling after 18 years study under in situ conditions, *Applied Geochemistry*. Elsevier. Vol. 84 (2017), 360-372.
- Vikman, Marjamaa, Nykyri, Small, Miettinen, Heikinheimo, Haavisto, Itävaara, 2018. The biogeochemistry of gas generation from low-level nuclear waste: Microbiological characterisation during 18 years study under in situ conditions, *Applied Geochemistry* 105, 55-67.
- Small, J., Vikman, M. 2021. Microbial impacts on gas production in LLW/ILW. In The Microbiology in Nuclear Waste Disposal, Eds. Jonathan R. Lloyd and Andrea Cherkouk, Elsevier, pp. 193-212.

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