KYT2022

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

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Gas generation in repository conditions

- 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 - - -> CO_2, CH_4, (H_2)

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

   Fe +2 H_2O → $Fe(OH)_2$ + H_2

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

Cellulose and hemicellulose are easily biodegradable

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Aim: To create disturbances in the gas generation experiment GGE 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)

Gas Generation Experiment (GGE)

- 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,but mixing of water may be induced by convection.
- Analyses during the experiment: Gas generation, composition of gas, water chemistry, key microbial groups
- Rather constant gas generation

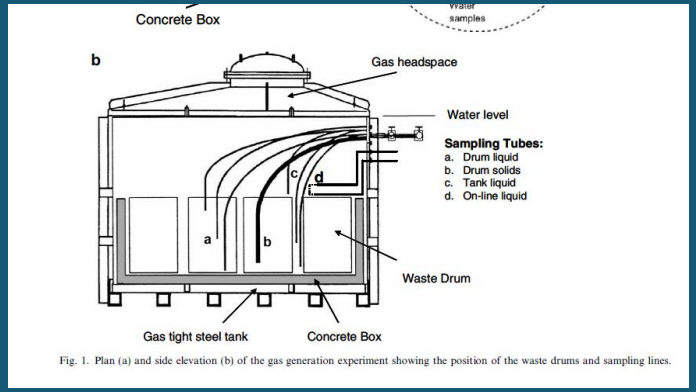
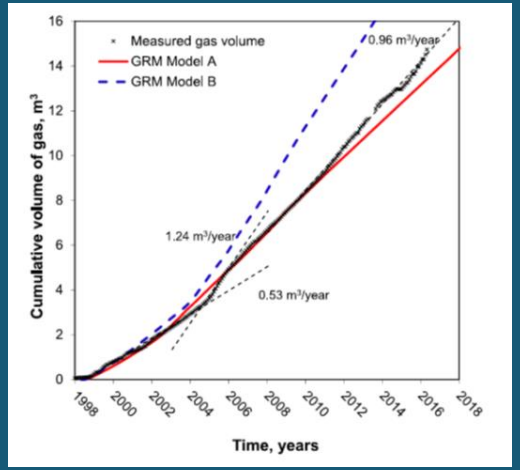


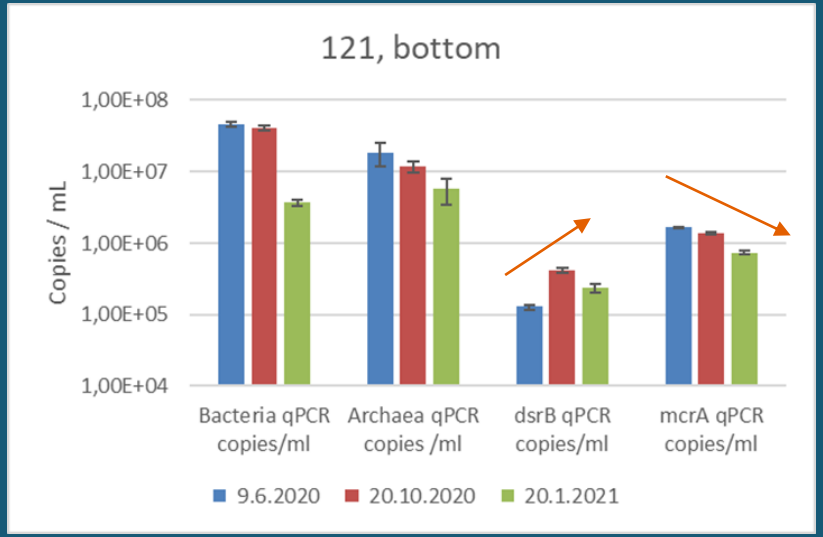
Fig. 1. Plan (a) and side elevation (b) of the gas generation experiment showing the position of the waste drums and sampling lines.



Figures: Small et al. 2017

Sulphate addition

- Changes in microbial population linked in methane generation detected
  - Decrease in the number of methanogens
  - Increase in the number of sulphate reducing bacteria
  - The diversity of methane-producing microbes stayed similar during the disturbance but certain sulphate reducing bacteria emerged to the microbial population in the tank water
- Sulphate reduction was not enhanced significantly
- No changes in gas generation and gas composition were detected

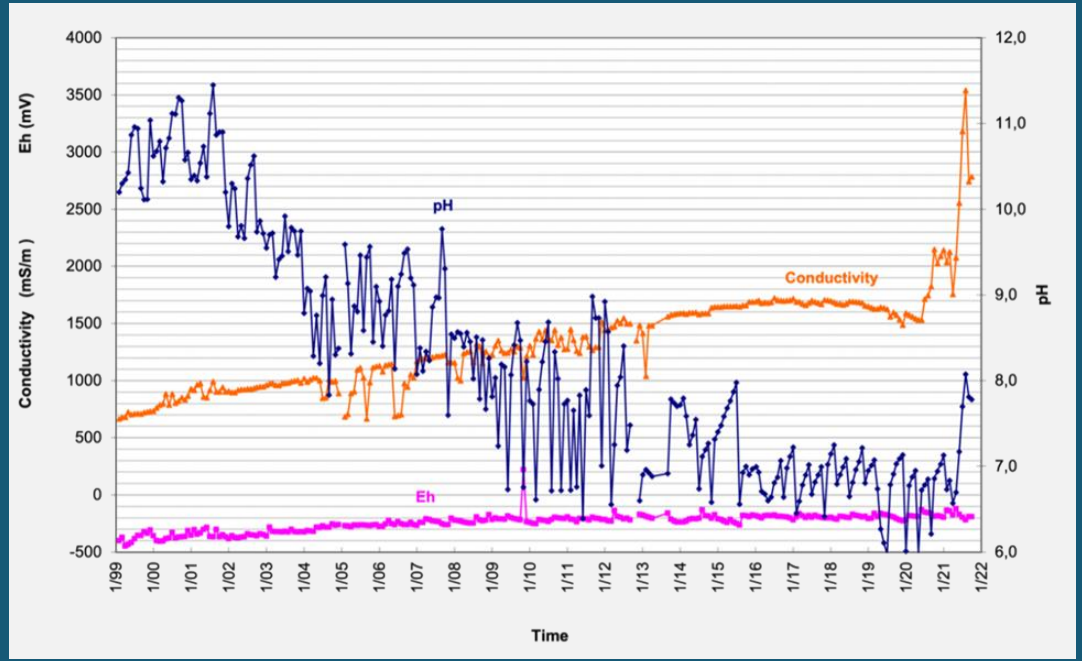


Sulphate addition Mixing

	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

Increase of tank water pH (6.7 to 8.1)

- Some changes in microbial population but no significant changes in methane producing microbes
- No changes in gas generation
- Some changes in gas composition were detected
  - CO_2 concentration in the released gas was reduced because CO_2 was partly absorbed into the more alkaline tank water
  - Soluble iron concentration in the tank water was reduced indicating precipitation of iron.

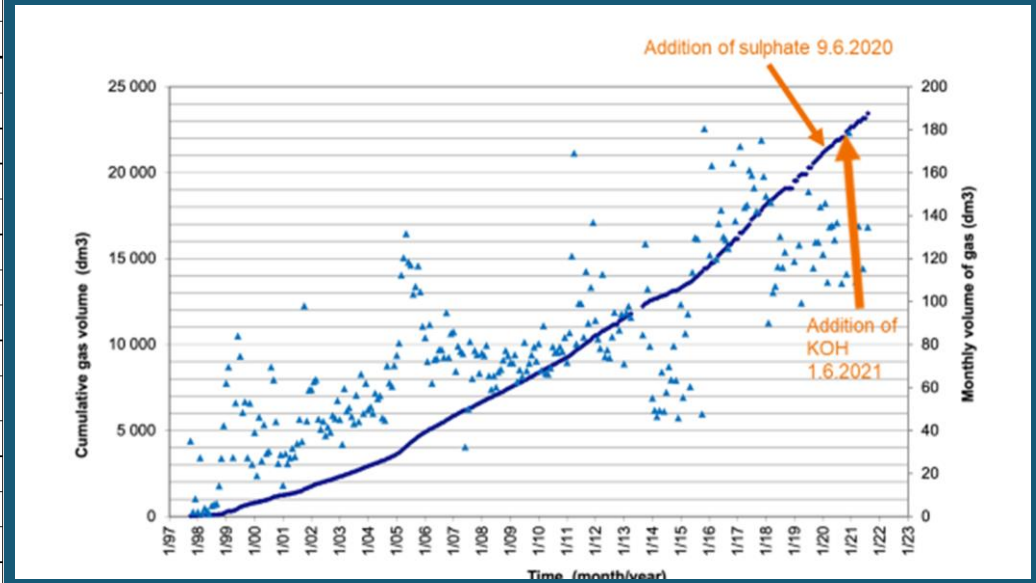


Gas evolution and gas composition

Sulphate addition →

KOH addition →

PVM	H_2 (t-%)	O_2 (t-%)	CH_4 (t-%)	CO_2 (t-%)	N2 (t.-%)
13.1.2020	<0,1	<0,1	90		0,48
15.5.2020	<0,1	<0,1	90		0,38
17.6.2020	0	1,7	82,9	10,4	5
8.7.2020	0	0,1	87,2	12,2	0,5
20.8.2020	<0,1	0,3	86,5	11,9	1,4
13.8.2020			87,5	10,2	
13.8.2020			89,2		
17.9.2020			86,8	10,04	
17.9.2020	<0,1	<0,1	90,3		<2,0
21.10.2020			87,7	9,99	
20.1.2021	<0,1	<0,1			<2,0
20.1.2021			87,5	10,07	
28.5.2021			88,6	11,1	
26.8.2021	<0,1	<0,1			2
26.8.2021			91	4,35	



- No significant changes in gas generation
- Gas composition: CO_2-concentration in released gas decreased from 11% to 4%

Conclusions

- The gas generation in the GGE was stable and neither the addition of sulphate nor increased pH influenced gas generation rate
- The obtained results can be used to verify modelling and to improve safety assessment related to the final disposal of low-level radioactive waste in Finland

Publications

- Small, J., Nykyri, M., Vikman, M., Itävaara, M., Heikinheimo, L., 2017. The biogeochemistry of gas generation from low-level nuclear waste: Modelling after 18 years study under in situ conditions, *Applied Geochemistry*. Elsevier. Vol. 84, 360-372.

- Vikman, Marjamaa, Nykyri, Small, Miettinen, Heikinheimo, Haavisto, Itävaara, 2019. 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. 2020. 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.

- Vikman, M., Sohlberg, E., 2022, The influence of chemical conditions on gas generation in the disposal of low level maintenance waste (KaMu), VTT Technical Research Centre of Finland. 23 p. VTT Research Report; No. VTT-R-00151-22.

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