

## Gas Generation and Transport in Subsurface Repository for LILW

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### 1. Introduction

Gases such as methane, hydrogen and etc have been generated in subsurface repository for low and intermediate level waste (LILW) due to metal corrosion, microbial degradation of soluble organics, and radiolysis. The gas generation will cause the pressure built-up in the repository, since the subsurface repository is constructed with concrete. The built-up pressure expels water from the repository. It also plays a role to increase the release of radionuclides to the environments.

Cementitious material, bentonite, gravel, sand, or their mixtures have been applied as backfill materials in different subsurface repositories. These materials work as engineered barrier systems (EBSs) to reduce water infiltration into the subsurface repository and to retard the migration of radionuclides. These materials significantly influence the gas transport in the repository. The pressure built-up depends on the critical gas pressure of backfill materials.

The objective of this research is to understand the gas generation and its consequence on pressure built-up in the subsurface repository with different backfill materials.

### 2. Repository Description and Waste Inventory

The design of the repository is schematically shown in Fig. 1. It comprises a concrete cylinder with a height of 35 m and an outer diameter of 23.6 m. The thickness of concrete wall is 0.6 m. Upper dome with a height of 15 m and outer diameter of 29.7 is placed on the concrete cylinder. Operational tunnel is connected to the upper dome. The repository will be finally closed by constructing concrete plug in the operation tunnel as shown in Fig. 1. Gas ventilation system will be installed through the concrete plug.

It is assumed that 16,700 steel drums of 200 L will be placed in the subsurface repository. Every drum is assumed to contain 45.6 kg of organics, 41.8 kg of mild steel, 42.6 kg of plastic, and 16.0 kg of others.

Radioisotopes and their radioactivity in each steel drum are listed in Table 1. These data are used in estimating the gas generation due to radiolysis.

### 3. Gas Generation

The gas generation due to the metal corrosion and the microbial decomposition of soluble organics was estimated using computer program GAMMON[1]. MAXH2 was also used to estimate the gas generation due to the radiolysis of plastics.

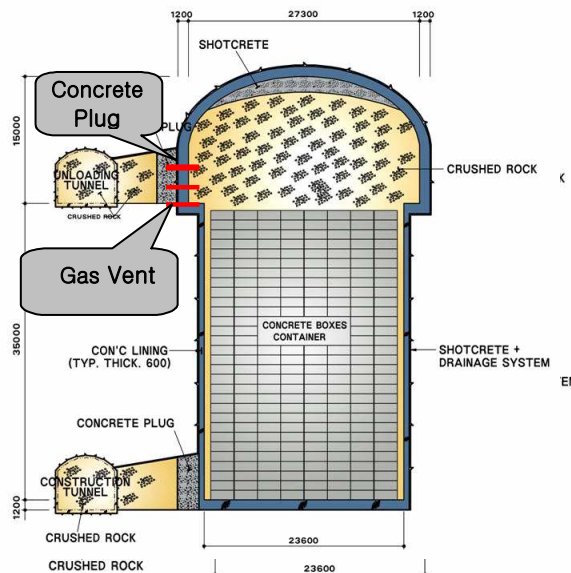


Fig. 1. Model repository for LILW equipped with gas vent system.

Table I: Radioisotopes and their activities used in MAXH2

Isotope	Radioactivity (Ci)
H-3	2.29E-02
C-14	9.11E-04
Fe-55	1.32E-01
Co-58	5.97E-02
Co-60	6.46E-03
Ni-59	3.38E-03
Ni-63	1.72E-02
Sr-90	1.31E-04
Nb-94	2.49E-04
Tc-99	8.59E-05
I-129	5.95E-06
Cs-137	1.32E-02
Ce-144	9.22E-03
Pu-239	2.55E-05

The estimated gas generation rate from the corrosion of metals is presented in Fig. 2. The gas generation rate was calculated for steel drums and mild steels in drums. During 1,000 years, hydrogen gas of  $9.0 \times 10^4 \text{ m}^3$  will be generated from corrosion of metals.

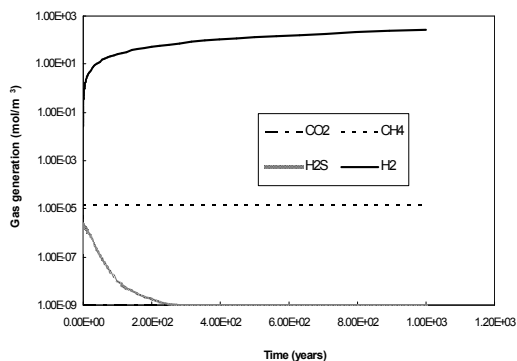


Fig. 2. Gas generation rate due to metal corrosion

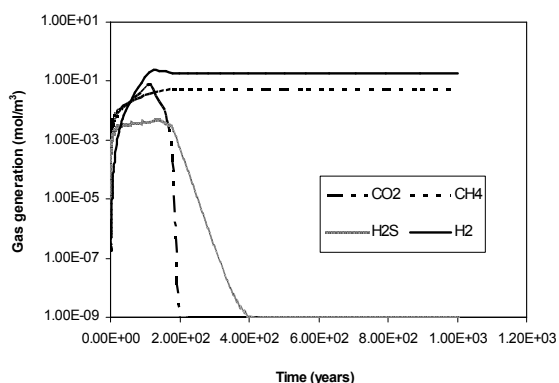


Fig. 3. Gas generation due to microbial degradation

So far, eight different microbial species have been considered for estimating the gas generation from the microbial decomposition of soluble organics. Initial microbial concentration was assumed to be  $1.0 \times 10^{-7}$  kg/m<sup>3</sup>. Methane and hydrogen gases were mainly generated from the microbial decomposition. After 1,000 years, their volume was  $6.38 \times 10^1$  and  $1.83 \times 10^1$  m<sup>3</sup>, respectively.

Plastics and rubbers were degraded by radiolysis, and consequently hydrogen gas is generated. Totally,  $4.22 \times 10^1$  m<sup>3</sup> of hydrogen gas will be generated after 1,000 years. However, the gas generation due to the radiolysis will be only 0.047 % of total gas volume which will be generated from metal corrosion. Therefore, it is acceptable to neglect the negligible gas generation from radiolysis.

#### 4. Gas Transport in the Repository

Gas transport through concrete structure is mainly controlled by the capillary pressure of the small pores in concrete. Generally, fresh concrete shows 1.5 MPa of critical gas pressure[2]. Hence, the pressure built-up takes place in the repository. This causes decrease of groundwater level in the repository, expels water in concrete pores, and finally damages concrete wall and other EBSs. This phenomenon enhances the release of radionuclide to the environment. Therefore, proper vent system should be equipped with the repository. In the

model repository, the gas vent system will be installed through the concrete plug, as shown in Fig. 1.

The gases dissolved in pore water also migrate through the concrete wall due to diffusion. The molecular diffusion is significantly affected by Henry's law constants and diffusion coefficients of the gases, and the tortuosity and porosity of the concrete. After the installation of gas vent, water level in the repository depends on the critical gas pressure of gas vent system. When the critical gas pressure of gas vent system is kept to 50 kPa, gas pressure in the repository is equilibrated with water level decreased to 5 m from the top. In this condition, the diffused gas dissolved in pore water occurs through the repository. For example, hydrogen concentration outside the repository is estimated to be  $1.07 \times 10^{-7}$  M to  $1.74 \times 10^{-9}$  M with the concrete tortuosity of 0.5 and 0.1 in 1,000 years, respectively using Fick's 2<sup>nd</sup> law. This result implies that the diffusion of hydrogen gas dissolved in pore water is not a dominant process in the transport of gases in the repository.

Backfill materials also influence the gas transport in the repository. Cementitious material has been considered as one of backfill candidates in the subsurface repository, because of its advantages including high pH level of pore water. However, it shows high critical gas pressure. Hence, it could cause significant pressure built-up in the repository. Porous mortar could be used as a backfill material instead of cementitious material to prevent the pressure built-up. Other backfill materials such as sands and gravels would not influence the gas transport in the repository because of their high pore size.

#### 5. Conclusion

The gas generation was estimated in the model repository which contained 16,700 steel drums of 200 L. The majority of gas generation resulted from the corrosion of metals. Consequently, the corrosion of metals generated  $9.0 \times 10^4$  m<sup>3</sup> of hydrogen gas after 1,000 years. The concrete wall exhibits 1.5 MPa of critical gas pressure. Hence, gas generation seriously damages the concrete structure and other EBS in the repository without the gas vent system. Backfill materials significantly influence the gas transport in the repository. Hence, porous mortar could be used as a backfill material in the repository instead of cementitious material because of its porous structure.

#### REFERENCES

- [1] P. J. Agg, Y. Gunn, D. Wibling, "GAMMON: A Computer Program Addressing Gas Generation in Radioactive Waste Repositories, Parts A-E, Nirex Report NSS/R338, 1997.
- [2] L. Moreno, "Project SAFE Gas related processes in SFR", SKB, R-01-11, 2001.